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Diffusion Coefficients of Ternary Mixtures of Water, Glucose, and Dilute Ethanol, Methanol, or Acetone by the Taylor Dispersion Method

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The Taylor dispersion technique is used to determine the diffusion coefficients of the ternary systems glucose + water + dilute methanol, ethanol, or acetone at 25 °C and up to a glucose mole fraction of 0.065. The dispersion of the injected solutes is recorded by a differential refractometer and an ultraviolet−visible detector. The diffusion coefficients are calculated directly by fitting the theoretical dispersion equations to about six experimental curves simultaneously. The precision of the diffusion coefficients is dependent on the relative detector sensitivities of the components. The determination of the main-diffusion coefficients is more precise than of the cross-diffusion coefficient (±2% vs ±5−10%).

Introduction

Multicomponent diffusion in liquids plays an important role in many chemical engineering processes such as distillation and extraction. For the analysis of the mechanism of volatile loss during the drying of food liquids, ternary diffusion data are required (Chandrasekaran and King, 1972). An experimental technique to measure multicomponent diffusion is the Taylor dispersion method (e.g., Leaist, 1991; van de Ven-Lucassen et al., 1997 and references therein). In a dispersion experiment, a slow, laminar flow of a liquid mixture is pumped through a long capillary tube and a narrow pulse of a mixture of a slightly different composition is injected into this tube. The injected solutes spread out owing to the combined effects of convective flow and molecular diffusion. At the end of the diffusion tube the dispersion is monitored by a flow-through detector (differential refractometer, ultraviolet−visible detector). The interdiffusion coefficients (called "diffusion coefficients" in this paper) are calculated by fitting the dispersion equations to the experimental curves. In this work the ternary diffusion coefficients are determined for ternary mixtures of α-D-glucose, water, and dilute ethanol, methanol, or acetone.

Theory

Diffusion in a three-component system solvent (0) + solute (1) + solute (2) is described by the coupled Fick equations

$$J_1 = -D_{11}\nabla C_1 - D_{12}\nabla C_2 \tag{1}$$

and

$$J_2 = -D_{21}\nabla C_1 - D_{22}\nabla C_2 \tag{2}$$

where J_i is the molar flux of component i and ∇C_i the gradient in the concentration of component i. The diffusion coefficient D_{ij} gives the flux of component i driven by the gradient in the concentration of component j; D_{ii} is called a main-diffusion coefficient and $D_{ij,i\neq j}$ a cross-diffusion coefficient.

To measure the ternary diffusion coefficients by the Taylor dispersion technique, a pulse of solution of composition $\bar{C}_{1,\mathrm{b}} + \Delta\bar{C}_1$, $\bar{C}_{2,\mathrm{b}} + \Delta\bar{C}_2$ is injected into the ternary mixture of composition $\bar{C}_{1,\mathrm{b}}$, $\bar{C}_{2,\mathrm{b}}$ flowing slowly through a long capillary tube. The pulse spreads out owing to the laminar velocity profile and molecular diffusion. The concentrations of the eluted solutes at the end of the diffusion tube are given by the fundamental working equations of Price (Price, 1988). If the concentration of component 2 tends to zero (component 2 is a tracer), it is impossible to produce a coupled flow of component 2 and $D_{21} \approx 0$; Price's equations simplify to (e.g., Leaist, 1991; van de Ven-Lucassen et al., 1997)

$$\bar{C}_1 = \frac{-\dfrac{D_{12}}{D_{11}-D_{22}}M_2}{2\pi R^2}\,\frac{1}{\sqrt{\pi\dfrac{R^2U^2}{48D_{22}}t}}\exp\left\{-\frac{(t-\tau)^2}{4\dfrac{R^2}{48D_{22}}t}\right\} + \frac{M_1 + \dfrac{D_{12}}{D_{11}-D_{22}}M_2}{2\pi R^2}\,\frac{1}{\sqrt{\pi\dfrac{R^2U^2}{48D_{11}}t}}\exp\left\{-\frac{(t-\tau)^2}{4\dfrac{R^2}{48D_{11}}t}\right\} \tag{3}$$

and

$$\bar{C}_2 = \frac{M_2}{2\pi R^2}\,\frac{1}{\sqrt{\pi\dfrac{R^2U^2}{48D_{22}}t}}\exp\left\{-\frac{(t-\tau)^2}{4\dfrac{R^2}{48D_{22}}t}\right\} \tag{4}$$

in which M_i represents the number of moles of component i in the injected pulse in excess of those in the same volume of the carrier stream. $\bar{C}_i$ is the radially averaged concentration of component i at time t relative to the background concentration $\bar{C}_{i,\mathrm{b}}$, τ is the mean residence time, R the internal radius of the diffusion tube, and U is the linear velocity averaged over the cross section. The assumptions made in the derivation of the working equations are similar

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to those of the binary dispersion technique with an effective diffusion coefficient D_{11} or D_{22} (Alizadeh et al., 1980).

When the dispersion is monitored, the relation between the detector signal $s(t)$ and the concentration against time curves (eqs 3, 4) is assumed to be

$$s(t) = w_1\bar{C}_1 + w_2\bar{C}_2 + a + bt + \epsilon(t) \tag{5}$$

in which w_i is the detector sensitivity of component i, a and b compensate for the detector drift (which is assumed to be linear in time), and $\epsilon(t)$ is noise. The signal is sampled with a sample interval of $\Delta t = 0.98$ s. The concentration against time curve of component 1 is dependent on the injected amount of components 1 and 2 and on the main-diffusion coefficients as well as the cross-diffusion coefficient (eq 3). The concentration against time curve of component 2 resembles a single Gaussian, only dependent on one main-diffusion coefficient and independent of the injected amount of component 1 (eq 4). Determination of the cross-diffusion coefficient D_{12} is only possible under certain conditions depending on the ratio of w_1/w_2, the ratio of the injected amounts M_1/M_2, the values of the diffusion coefficients, and the noise level (van de Ven-Lucassen et al., 1997).

Diffusion coefficients can be calculated from the measured dispersion profiles in two different ways: calculation from the temporal moments (Leaist et al., 1993) or fitting of the theoretical eqs 3 and 4 to the experimental curve (e.g., Leaist and Hao, 1994). In this work only fitting procedures will be used with the diffusion coefficients, the mean residence time τ, and the detector sensitivities w_1 and w_2 as fitting parameters. In these fitting procedures the discrete signal s without the drift is required: $s = s^* - (a + bt)$, in which s^* is the discrete output signal of the detector. For the calculation of a and b, regions of the detected signal before and after the eluted solute peak, where the concentration is negligible, are visually marked in our software as baseline. Through these regions of the signal the drift is fitted (using a least-squares method) and subtracted from the signal (van de Ven-Lucassen et al., 1995).

Equipment and Experimental Procedure

An extensive description of the equipment for the measurement of diffusion coefficients in binary and ternary liquid systems is given by van de Ven-Lucassen et al. (1995, 1997). Solutions were prepared by mass and mixing, and degassed by sparging with helium. Injection solutions were made by volumetrical mixing of the degassed materials. To prevent bubbles from disturbing the flow, an in-line degasser (Separations DG1300) was installed. The HPLC pump (type LKB2150), which maintained a steady flow, was connected to an autosampler (Spark Marathon) with a fixed volume sample loop of 20 μL. Zero dead volume fittings were used to connect the diffusion tube with the autosampler and the ultraviolet−visible detector (UV detector) and a short capillary tube between the UV detector and the differential refractometer (RI detector). The diffusion tube was a 25-m length of PEEK (Poly Ether Ether Ketone) tubing wrapped in a 0.40-m diameter coil. The internal radius of the tube (0.52 ± 0.01 mm) was determined by gravimetry and residence time measurements. From diffusion measurements of the systems methanol + water (whole concentration range), ethanol + water, and acetone + water the internal radius was also calculated (data used were given in van de Ven-Lucassen et al., 1995, and references therein); this radius was not significantly different (0.521 ± 0.003 mm).

The eluted peaks were detected with the UV detector (Applied Biosystems 785A) and subsequently by the differential refractometer (Shodex SE61). The analogue output signal of the refractometer was converted by a Multilab system (A/D−D/A conversion system developed at the Eindhoven University of Technology). For the conversion of the analogue output signal of the UV detector, a hardware interface device (Strawberry-Tree Mini16 ADC) was built in the personal computer (PC). The multilab was used to interface between PC and RI detector and between PC and selection valve. The pump and the autosampler were controlled directly by the PC. Software has been developed for data acquisition and controlling the equipment as well as for processing of the data.

The procedure for the measurement of diffusion coefficients in binary systems is described in detail by van de Ven-Lucassen et al. (1995). Before a ternary experiment was started, the system was flushed for at least 6 h at the flow rate of the diffusion experiment to attain a stable, linear baseline in each detector. The flow velocity was set in accordance with the conditions, under which Price's equations 3 and 4 are valid (typically 0.12 $\mathrm{cm^3{\cdot}min^{-1}}$). Diffusion samples were injected every $1^1/_2$ − 2 h. After application of a baseline correction to each experimental curve, binary fits were done for dispersion peaks obtained from $M_2 = 0$ or $M_1 = 0$ injections: the mean residence time τ, the binary diffusion coefficient (D_{11} or D_{22}), and the peak area were calculated. The detector sensitivity (w_1 or w_2) was determined from the peak area and the injected amount. These values could be used as starting values for the fitting parameters in the ternary procedures. The fitting procedures are based on the nonlinear least-squares approximation between the experimental data points and the points calculated according to eqs 3 and 4 (van de Ven-Lucassen et al., 1997). These ternary fitting procedures are written in the SAS for Windows package (version 6.10, SAS Institute Inc.), using the method of Marquardt. They are able to fit several experimental curves simultaneously with the following fitting parameters:

(1) D_{11}, D_{22}, D_{12}, τ, w_1, and w_2

(2) D_{12}, τ, w_1, and w_2; D_{11} calculated from the binary diffusion experiments; D_{22} calculated from the UV signals (only for acetone as a tracer). Other combinations of fitting parameters and calculated parameters are possible.

Experimental Results and Discussion

Diffusion coefficients were measured for the binary systems methanol + water, ethanol + water, acetone + water, and glucose + water and for the ternary systems glucose + ethanol + water, glucose + methanol + water, and glucose + acetone + water.

Water deionized and filtered through a Milli-Q water purification system (Millipore, resistivity 18 MΩ·cm) was used. Analytical grade methanol (purity ≥ 99.8%, water < 0.05%), ethanol (purity ≥ 99.8%, water < 0.2%), acetone (purity ≥ 99.5%, water < 0.2%), and D(+)-glucose anhydrous (purity ≥ 99.0%, water < 0.2%) were obtained from Merck and used without further purification. All experiments were performed at 25 °C.

Binary Systems. Experiments were performed to show that the detector response was linear with concentration and to study the influence of the concentration of the injected sample on the measurement of the diffusion coefficients. Samples of increasing or decreasing glucose

Table 1. Diffusion Coefficients and Detector Sensitivities at 25 °C of the Systems Water (0) + Glucose (1) + Tracer (2)

mole fraction x_1	binary system water + glucose $D_{11,\mathrm{bin}}/10^{-10}\ \mathrm{m^2 \cdot s^{-1}}$	ternary system water (0) + glucose (1) + acetone (2) $D_{22,\mathrm{UV}}/10^{-10}\ \mathrm{m^2 \cdot s^{-1}}$
0	6.9	12.7
0.015	5.79	9.6
0.025	5.2	
0.035	4.58	6.56
0.05	3.80	4.92
0.065	3.14	3.62

Ternary Systems Water (0) + Glucose (1) + Tracer (2)

$x1$	$D_{11}/10^{-10}$ $\mathrm{m^2 \cdot s^{-1}}$	$D_{22}/10^{-10}$ $\mathrm{m^2 \cdot s^{-1}}$	$D_{12}/10^{-10}$ $\mathrm{m^2 \cdot s^{-1}}$	$w_1/10^4$ $\mathrm{RIU \cdot mol^{-1}}$	$w_1/10^4$ $\mathrm{RIU \cdot mol^{-1}}$
			Tracer Acetone (2)		
0.000	(7.05)[a]	(12.7)[a]	−0.004	3.474	0.479
0.015	5.75	9.95	0.18	3.380	0.550
0.025	5.36	8.31	0.29	3.105	0.559
0.035	(4.50)[a]	(6.56)[a]	0.51	3.237	0.588
0.050	3.78	5.02	0.54	3.110	0.362
0.65	3.11	3.84	0.57	2.990	0.633
			Tracer Ethanol (2)		
0.000	(7.05)[a]	(12.2)[a]	−0.016	3.449	0.360
0.015	5.75	9.34	0.31	3.389	0.425
0.025	5.11	7.98	0.59	3.209	0.376
0.035	4.44	6.55	0.60	3.201	0.435
0.050	3.77	4.91	0.69	3.111	0.282
0.065	3.11	3.69	0.64	2.990	0.492
			Tracer Methanol (2)		
0.000	(7.05)[a]	(15.3)[a]	−0.015	3.435	0.079
0.015	5.77	12.1	0.071	3.409	0.158
0.025	5.27	10.3	0.07	3.075	0.181
0.035	4.56	8.5	0.21	3.259	(0.108)[a]
0.050	3.77	6.43	0.49	3.111	0.002
0.065	3.11	4.77	0.29	2.990	0.256

[a] Not fitted; fixed values used in the fitting procedures and obtained from the binary experiments.

Table 2. Imprecision of the Diffusion Coefficients and Detector Sensitivities at 25 °C of the Systems Water (0) + Glucose (1) + Tracer (2)

mole fraction x_1	binary system water + glucose $D_{11,\mathrm{bin}}/\%$	ternary system water (0) + glucose (1) + acetone (2) $D_{22,\mathrm{UV}}/\%$
0	1.6	0.5
0.015	1.0	1.3
0.025	2.5	
0.035	1.7	0.8
0.05	1.2	1.2
0.065	1.0	1.3

Ternary Systems Water (0) + Glucose (1) + Tracer (2)

x_1	$D_{11}/\%$	$D_{22}/\%$	$D_{12}/\%$	$w_1/\%$	$w_2/\%$
			Tracer Acetone (2)		
0.000			49	0.02	0.06
0.015	0.08	0.5	6.5	0.04	0.08
0.025	0.19	1.0	11	0.08	0.15
0.035			0.1	0.02	0.03
0.050	0.21	1.0	9.3	0.09	0.32
0.065	0.08	0.2	2.9	0.04	0.06
			Tracer Ethanol (2)		
0.000			6.0	0.01	0.04
0.015	0.04	0.2	2.1	0.02	0.04
0.025	0.31	1.2	7.8	0.14	0.32
0.035	0.05	0.1	0.8	0.02	0.03
0.050	0.21	0.8	7.5	0.09	0.37
0.065	0.15	0.4	5.8	0.06	0.13
			Tracer Methanol (2)		
0.000			6.5	0.01	0.16
0.015	0.03	0.5	4.8	0.01	0.09
0.025	0.16	1.3	15	0.07	0.25
0.035	0.28	1.8	10	0.12	
0.050	0.20	1.3	6.0	0.09	54
0.065	0.16	0.8	5.4	0.07	0.23

concentration were injected into binary mixtures of glucose and water, and the peak area and the diffusion coefficient were calculated. For all values of the injected excess amount of glucose, the detector response was linear. Below an absolute value of the injected excess of 1 × 10^{-5} mol, the diffusion coefficient was independent of the injected amount; at higher values the diffusion coefficient increased, probably owing to secondary flow effects. Therefore, these high injection concentrations were not used for the determination of the binary and ternary diffusion coefficients. Injections of solutions of increasing methanol, ethanol, or acetone concentrations into pure water showed also a linear detector response and an independence of the concentration difference up to 4−6 vol % between injection sample and solvent. Injections of solutions of acetone in glucose−water mixtures were detected also by the UV detector at a wavelength of 266 nm. At this wavelength only acetone was detected; i.e., $w_1 = 0$ in eq 5. From the UV signal the diffusion coefficient D_{22} ($D_{22,\mathrm{UV}}$) and the peak area were calculated. The detector response was linear with the injected amount of acetone, and the diffusion coefficient was independent of the injected amount of acetone.

The diffusion coefficients of the binary system glucose + water (D_{11}) and of the tracer acetone in the glucose + water mixtures ($D_{22,\mathrm{UV}}$) are listed in Table 1; the precision of the results is given in Table 2. The values of the binary D_{11} are the mean of seven injections and the values of the $D_{22,\mathrm{UV}}$ are the mean of six injections, each with a different concentration. The confidence limits of the binary D_{11} and $D_{22,\mathrm{UV}}$ ("precision") were calculated according to the Student's t distribution, probability level 95%, two-tail test (Perry and Chilton, 1969).

Ternary Systems. The ternary diffusion coefficients of the systems glucose + acetone + water, glucose + ethanol + water, and glucose + methanol + water were determined by injecting ternary mixtures into the binary glucose + water mixture. Injection samples were prepared in vials to give seven different values of M_1 with $\mathrm{M}_2 = 0$, and for each tracer component three different values of M_2 with $M_1 = 0$ and three different combinations of M_1 and M_2. The ternary diffusion coefficients D_{11}, D_{22}, and D_{12}, the mean residence time τ, and the detector sensitivities w_1 and w_2 were calculated by simultaneously fitting of six RI peaks, each with a different value of M_1 and M_2 (mostly two peaks with $M_1 = 0$, two peaks with $M_2 = 0$, and two combination peaks). An estimation of the standard errors of all parameters was given by the nonlinear SAS fitting procedures used, and a 95% confidence interval was calculated ("precision"). For all mixtures, the main-diffusion coefficients were more precise than the cross-diffusion coefficient. The inaccuracy of the residence time τ was less than 0.005%. There was a strong correlation between the cross-diffusion coefficient D_{12} and the main-diffusion coefficient of the tracer D_{22}. The values of the diffusion coefficients obtained from a different group of six peaks agreed within 1−2% for the main-diffusion coefficients and within the precision of the fitting procedure for the cross-diffusion coefficients. No significant difference was observed between the calculated diffusion coefficients if the starting values were changed, provided convergence was achieved. In general, experiments with tracer component methanol were less accurate. The dispersion curves had a poorer signal-to-noise ratio, and convergence of the fitting procedures was harder

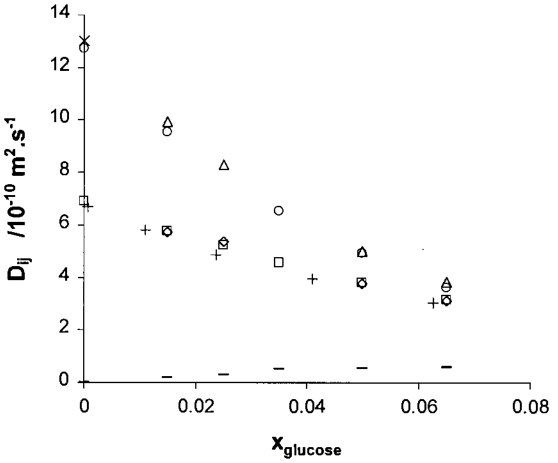

Figure 1. Ternary diffusion coefficients of the system glucose (1) + acetone (2) + water (0) at 25 °C and $x_2 = 0$: (□) binary D_{11}, (◇) ternary D_{11}, (△) ternary D_{22}, (−) ternary D_{12}, (○) $D_{22,UV}$, this work; (+) binary D_{11}, Gladden and Dole (1953), Uedaira and Uedaira (1985); (×) binary D_{22}, Tyn and Calus (1975).

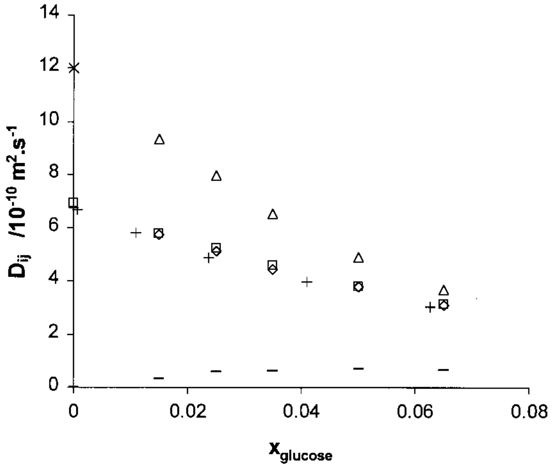

Figure 2. Ternary diffusion coefficients of the system glucose (1) + ethanol (2) + water (0) at 25 °C and $x_2 = 0$: (□) binary D_{11}, (◇) ternary D_{11}, (△) ternary D_{22}, (−) ternary D_{12}, this work; (+) binary D_{11}, Gladden and Dole (1953), Uedaira and Uedaira (1985); (×) binary D_{22}, van de Ven−Lucassen et al. (1995).

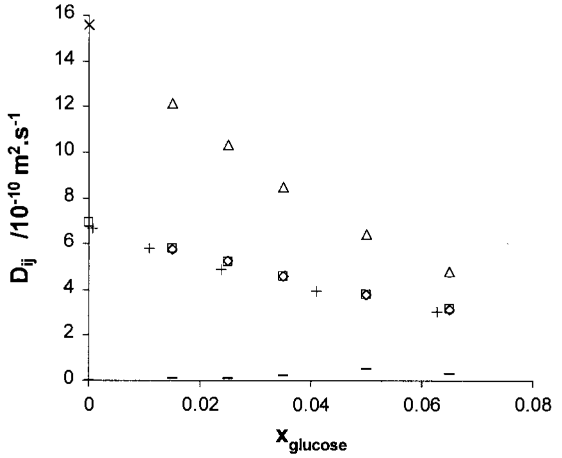

Figure 3. Ternary diffusion coefficients of the system glucose (1) + methanol (2) + water (0) at 25 °C and $x_2 = 0$: (□) binary D_{11}, (◇) ternary D_{11}, (△) ternary D_{22}, (−) ternary D_{12}, this work; (+) binary D_{11}, Gladden and Dole (1953), Uedaira and Uedaira (1985); (×) binary D_{22}, van de Ven−Lucassen et al. (1995).

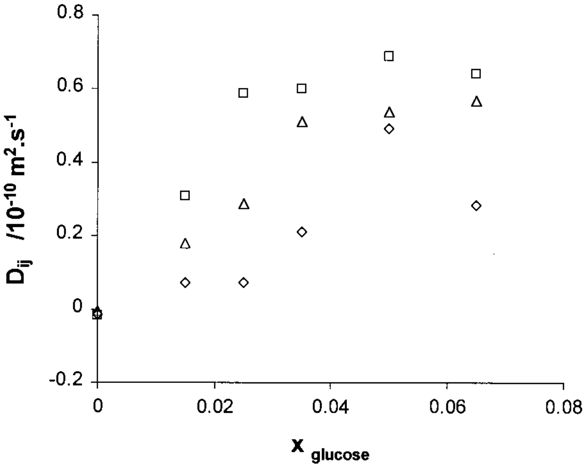

Figure 4. Cross-diffusion coefficients D_{12} of the system glucose (1) + tracer (2) + water (0) at 25 °C and $x_2 = 0$: (□) ethanol, (◇) methanol, (△) acetone.

to achieve (e.g., at $x_1 = 0.035$, $x_1 = 0.050$). This was caused by the lower RI sensitivity of methanol. The strong correlation between D_{12} and D_{22} was studied by fitting six RI peaks (at $x_1 = 0.065$) simultaneously with D_{11} and D_{22} fixed at a value with a deviation of ±5% of the D_{11} and D_{22}, calculated in a previous fitting procedure. The value of the cross-diffusion coefficient D_{12}, calculated with the fixed D_{11} and D_{22}, was within the confidence interval of the value of the D_{12}, calculated in the fitting procedure of D_{11}, D_{22}, and D_{12} simultaneously. Results of the ternary diffusion measurements are listed in Tables 1 and 2 and shown in Figures 1−4. The binary D_{11} and the $D_{22,UV}$ of acetone agreed well with the values calculated by the ternary fitting procedures.

Comparison of the values at infinite dilution ($x_1 = 0$) and of the binary D_{11} with literature values in Figures 1−3 showed a good agreement. Chandrasekaran and King (1972) determined the ternary diffusion coefficients for the system ethanol (1, tracer) + water (2) + glucose (0). As the four diffusion coefficients are dependent on the choice of the solvent, the values of Chandrasekaran and King (1972) had to be converted to the system glucose (1) + ethanol (2, tracer) + water (0). Equations used for this conversion were (Vitagliano et al., 1978) $D_{21} \approx 0$, $D_{12}^* \approx 0$, $D_{11} = D_{22}^*$, $D_{22} = D_{11}^*$, and $D_{12} = (V_2/V_1)D_{22}^* - (\bar{V}_0/V_1)D_{21}^* - (V_2/V_1)D_{11}^*$, in which V_i is the partial molar volume of component i and D_{ij}^* are the original diffusion coefficients, determined by Chandrasekaran and King (1992). The partial molar volumes were calculated using equations describing the partial molar volumes as a function of the mole fractions, the molar mass, and the (mole fraction derivatives of the) density of the mixtures. Density values were obtained from Cerdeirina et al. (1997) and from Taylor and Rowlinson (1955). Comparison of the converted values with the measured values in Figure 5 shows a good agreement for the main-diffusion coefficient D_{11}. The values of the measured main-diffusion coefficient D_{22} and of the measured cross-diffusion coefficient D_{12} are higher than the converted literature values. The accuracy of the converted cross-diffusion coefficient was low owing to the procedure followed. The estimation of D_{21}^* and D_{11}^* from a logarithmic graph occurred with only a moderate precision (imprecision > 5%). Chandrasekaran and King showed the results in logarithmic graphs of the diffusion coefficients versus the concentration of water (and the weight percent sugar in solution) and did not mention the accuracy of the measurements explicitly. The calculation using the partial molar volumes and the estimated diffusion coefficients decreased

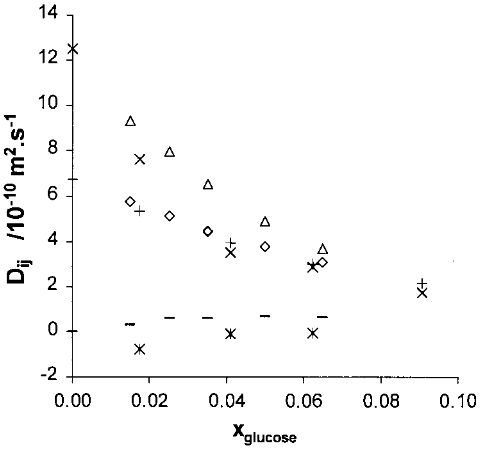

Figure 5. Comparison of the experimental ternary diffusion coefficients of the system glucose (1) + ethanol (2) + water (0) with previous work at 25 °C and $x_2 = 0$: (◇) ternary D_{11}, (△) ternary D_{22}, (−) ternary D_{12}, this work; (+) ternary D_{11}, (×) ternary D_{22}, (*) ternary D_{12}, Chandrasekaran and King (1972).

the precision. Furthermore, the cross-diffusion coefficient D^*_{12} was not set equal 0. It is therefore understandable that comparison of the measured D_{12} with the converted D_{12} does not show good agreement.

Conclusions

The Taylor dispersion method is a fast and convenient technique for measuring diffusion coefficients in liquid systems. In ternary systems with one component infinitely diluted, the precision of the method is dependent on the relative detector sensitivities of the components. Of the systems studied in this paper, the measured main-diffusion coefficients D_{11} and D_{22} are more precise than the measured cross-diffusion coefficient D_{12}.

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