Experimental Distinct Diffusion Data for 14 Binary Nonelectrolyte Mixtures

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In a recent publication, Mills *et al.* (*1*) calculated distinct diffusion coefficients in 4 frames of reference for 14 binary nonelectrolyte mixtures. Because of space limitations, some of the data were represented graphically in that paper. The distinct diffusion coefficient data for the mass-, volume- and number-fixed frames of reference for all systems are given numerically here. The thermodynamic factor $(\partial \ln a_i/\partial \ln x_i)_{T,P}$, which is often used in mass-transport theories, is also given.

Introduction

In the dynamical situation, quantitative knowledge of the interactions between molecules of the same species in nonelectrolyte mixtures requires the calculation of generalized transport coefficients. A very useful coefficient of this kind is the distinct diffusion coefficient, D_{ij}^{d}. Distinct diffusion coefficients are important because their definition in terms of velocity correlation functions links them directly to statistical mechanics. They also have the advantage of finite limiting values and an inherent symmetry. In a recent publication Mills *et al.* (*1*) described the calculation of experimentally-derived D_{ij}^{d} in various frames of reference for 14 nonelectrolyte mixtures and used them to assess the suitability of a particular reference frame for discussing macroscopic solution properties. That paper and the present one are complementary.

In the mass-fixed (or barycentric) frame of reference the $(D_{ij}^{d})^{M}$ can be calculated from the following equations:

$$(D_{ij}^{d})^{M} = -D^{V}M_iM_j/((x_iM_i + x_jM_j)^2B_i^{x}) \qquad i \neq j \quad (1)$$

$$(D_{ii}^{d})^{M} = D^{V}M_j^2x_j/((x_i(x_iM_i + x_jM_j)^2B_i^{x})) - ((D_i^{s})_j/x_i) \quad (2)$$

where D^{V} is the interdiffusion (mutual) coefficient on the volume-fixed frame of reference, $(D_i^{s})_j$ is the intradiffusion (self) coefficient of i in a mixture of i and j, B_i^{x} is the thermodynamic factor $(\partial \ln a_i/\partial \ln x_i)_{T,P}$ with concentrations expressed in mole fractions x_i, and M_i is the molecular weight of species i. The defining equations (given in ref 1) for the other frames are essentially similar to eqs 1 and 2 with respect to experimental variable content.

Thus, the primary pieces of data needed for the calculation of the D_{ij}^{d} in binary mixtures are the interdiffusion and intradiffusion coefficients. The thermodynamic factor $(\partial \ln a_i/\partial \ln x_i)_{T,P}$ is itself a secondary piece of data and is in turn derived from primary experimental data such as vapor–liquid equilibria and excess enthalpy measurements.

The probable errors in the $[D_{ij}^{d}]^{R}$ are not easily quantified. There is a small error in the calculation of the $[D_{ij}^{d}]^{V}$ which arises from the use of partial molar volumes, but as these are determined from density measurements, this error will be negligible compared to others. The accuracy of the primary diffusion data is reasonably good, and most of the error is associated with the precision of the method of measurement. Intradiffusion coefficients measured in diaphragm cells generally will have errors in the range ±0.2–0.5%, those from spin-echo NMR in the range ±1–4%, and those by capillary techniques *ca.* ±1%. Interdiffusion coefficients as measured by optical methods (Gouy, Raleigh, Mach-Zehnder) generally have errors in the range ±0.1–0.5%, while for those from diaphragm cells, the range is ±0.5–1%. The error in the thermodynamic factor $[\partial \ln a_i/\partial \ln x_i]_{T,P}$ (denoted by B_i^{x} in eqs 1 and 2) is difficult to determine because it is a secondary quantity with two derivatives being involved in its calculation. For ideal systems the error in B_i^{x} would be *ca.* ±1%, but for nonideal systems (such as those with water as a component) it could be several percent. For these latter systems the error in B_i^{x} will obviously dominate the total error.

It should be remarked that although, on average, the cumulative error in the $[D_{ij}^{d}]^{R}$ may well be more than ±1%, these values are still very useful for the macroscopic, qualitative description of solution properties and for comparison with quantities derived from present theory (*1*).

For the sake of uniformity, we have quoted all $[D_{ij}^{d}]^{R}$ to two decimal places in the range $(0–5) \times 10^{-9}$ m^2 s^{-1} and to one decimal place in the range 5–10 and have omitted decimal places for values greater than 10. $[\partial \ln a_i/\partial \ln x_i]_{T,P}$ values are customarily quoted to three or four decimal places. They are given here to three decimal places although, as a measure of their accuracy, this would be an overestimate in many cases.

In order for the reader to gain some appreciation of the overall error in the $[D_{ij}^{d}]^{R}$ for each *system*, the following code is used in Table 1. For the type of measurement, S = intradiffusion, I = interdiffusion, and B refers to the thermodynamic data. The symbol a is used for the diaphragm-cell method, b for spin-echo NMR, and c for capillary measurements. Optical methods are denoted by d. The source of data is then given as the number of the reference. Thus, for the system cyclooctane (1) + cyclopentane (2) the code is S = a,b,2; I = d,2; B = 3. The original source of data can then be consulted if a more detailed knowledge of the error is needed.

It will also be noted that occasionally there are blank spaces in the $x_1 = 0.1–0.2$ region (for either component) in some of the nonideal systems. This is due to the

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Table 1. Experimental Distinct Diffusion Coefficients of Binary Nonelectrolyte Systems

	mass-fixed			volume-fixed			number-fixed			
x_1	$D^d_{11}/(10^{-9}$ $m^2 \cdot s^{-1})$	$D^d_{12}/(10^{-9}$ $m^2 \cdot s^{-1})$	$D^d_{22}/(10^{-9}$ $m^2 \cdot s^{-1})$	$D^d_{11}/(10^{-9}$ $m^2 \cdot s^{-1})$	$D^d_{12}/(10^{-9}$ $m^2 \cdot s^{-1})$	$D^d_{22}/(10^{-9}$ $m^2 \cdot s^{-1})$	$D^d_{11}/(10^{-9}$ $m^2 \cdot s^{-1})$	$D^d_{12}/(10^{-9}$ $m^2 \cdot s^{-1})$	$D^d_{22}/(10^{-9}$ $m^2 \cdot s^{-1})$	$[\partial \ln a_i/\partial \ln x_i]_{T,P}$
(a) Cyclooctane (1) + Cyclopentane (2) (T = 298.15 K; S = a,b,2; I = d,2; B = 3)										
0	−4.31	−3.78	−3.11	−3.40	−3.32	−3.11	−1.48	−2.36	−3.11	1.000
0.1	−3.44	−3.02	−2.45	−2.81	−2.76	−2.56	−1.33	−2.11	−2.75	0.993
0.2	−2.83	−2.43	−2.01	−2.33	−2.30	−2.16	−1.20	−1.91	−2.50	0.993
0.3	−2.21	−1.97	−1.56	−1.93	−1.92	−1.74	−1.08	−1.72	−2.18	0.995
0.4	−1.80	−1.61	−1.22	−1.61	−1.62	−1.41	−0.99	−1.55	−1.91	1.001
0.5	−1.46	−1.32	−0.96	−1.34	−1.36	−1.14	−0.89	−1.40	−1.68	1.006
0.6	−1.18	−1.09	−0.77	−1.11	−1.14	−0.94	−0.80	−1.26	−1.49	1.008
0.7	−0.97	−0.90	−0.56	−0.93	−0.96	−0.72	−0.73	−1.13	−1.27	1.007
0.8	−0.82	−0.74	−0.39	−0.79	−0.80	−0.54	−0.68	−1.01	−1.08	1.005
0.9	−0.65	−0.61	−0.23	−0.64	−0.67	−0.36	−0.59	−0.89	−0.88	1.002
1.0	−0.54	−0.50	−0.09	−0.54	−0.55	−0.21	−0.54	−0.79	−0.69	1.000
(b) Chlorobenzene (1) + Benzene (2) (T = 298.15 K; S = a,4; I = d,4; B = 5)										
0	−2.59	−3.05	−2.21	−1.34	−2.42	−2.21	−0.73	−2.12	−2.21	1.000
0.1	−2.68	−2.88	−1.94	−1.62	−2.42	−2.10	−1.07	−2.18	−2.16	0.965
0.2	−2.74	−2.63	−1.71	−1.88	−2.34	−1.99	−1.40	−2.16	−2.11	0.960
0.3	−2.72	−2.38	−1.51	−2.03	−2.23	−1.89	−1.63	−2.12	−2.07	0.965
0.4	−2.61	−2.16	−1.35	−2.07	−2.12	−1.81	−1.75	−2.07	−2.04	0.972
0.5	−2.46	−1.96	−1.21	−2.06	−2.02	−1.73	−1.79	−2.03	−2.01	0.979
0.6	−2.31	−1.79	−1.10	−2.01	−1.93	−1.67	−1.81	−1.99	−1.99	0.985
0.7	−2.15	−1.65	−1.00	−1.95	−1.85	−1.61	−1.81	−1.96	−1.97	0.990
0.8	−2.01	−1.52	−0.91	−1.89	−1.78	−1.56	−1.80	−1.93	−1.96	0.994
0.9	−1.88	−1.41	−0.84	−1.82	−1.71	−1.50	−1.78	−1.90	−1.94	0.997
1.0	−1.76	−1.30	−0.77	−1.76	−1.64	−1.45	−1.76	−1.87	−1.92	1.000
(c) Cyclohexane (1) + Benzene (2) (T = 298.15 K; S = a,6; I = d,7; B = 7)										
0	−1.06	−2.24	−2.21	−1.78	−2.62	−2.21	−0.74	−2.08	−2.21	1.000
0.1	−1.17	−2.36	−2.22	−1.84	−2.63	−2.14	−0.92	−2.22	−2.25	0.890
0.2	−1.43	−2.43	−2.21	−1.97	−2.61	−2.06	−1.15	−2.32	−2.28	0.817
0.3	−1.61	−2.45	−2.17	−2.04	−2.55	−1.96	−1.36	−2.38	−2.28	0.774
0.4	−1.71	−2.43	−2.10	−2.06	−2.47	−1.84	−1.50	−2.40	−2.25	0.752
0.5	−1.76	−2.38	−2.00	−2.03	−2.36	−1.69	−1.58	−2.38	−2.18	0.751
0.6	−1.76	−2.30	−1.86	−1.95	−2.23	−1.52	−1.62	−2.33	−2.07	0.767
0.7	−1.71	−2.19	−1.71	−1.85	−2.07	−1.35	−1.62	−2.25	−1.94	0.800
0.8	−1.64	−2.05	−1.54	−1.72	−1.91	−1.19	−1.58	−2.15	−1.79	0.849
0.9	−1.55	−1.90	−1.39	−1.58	−1.75	−1.06	−1.52	−2.02	−1.65	0.916
1.0	−1.44	−1.74	−1.27	−1.44	−1.58	−0.98	−1.44	−1.87	−1.53	1.000
(d) Octamethylcyclotetrasiloxane (1) + Benzene (2) (T = 298.15 K; S = a,b,8,9; I = d,10; B = 11)										
0	−4.34	−4.37	−2.21	−3.63	−4.01	−2.21	2.10	−1.15	−2.21	1.000
0.1		−2.99	−1.08		−2.88	−1.22		−1.29	−2.20	0.803
0.2	−2.23	−2.05	−0.54	−2.06	−2.04	−0.70	0.85	−1.31	−2.16	0.806
0.3	−1.77	−1.45	−0.29	−1.67	−1.48	−0.44	0.35	−1.29	−2.10	0.860
0.4	−1.36	−1.07	−0.16	−1.31	−1.11	−0.29	0.11	−1.26	−2.03	0.913
0.5	−1.07	−0.81	−0.10	−1.04	−0.85	−0.21	−0.05	−1.23	−1.96	0.955
0.6	−0.87	−0.64	−0.06	−0.86	−0.67	−0.16	−0.18	−1.20	−1.88	0.984
0.7	−0.70	−0.51	−0.02	−0.69	−0.55	−0.10	−0.25	−1.18	−1.80	1.000
0.8	−0.51	−0.42	0.03	−0.51	−0.45	−0.05	−0.25	−1.16	−1.71	1.004
0.9	−0.43	−0.35	0.07	−0.43	−0.38	0.01	−0.32	−1.13	−1.61	1.004
1.0	−0.38	−0.29	0.11	−0.38	−0.31	0.06	−0.38	−1.09	−1.50	1.000
(e) n-Hexane (1) + Benzene (2) (T = 298.15 K; S = a,4; I = d,4; B = 4,5)										
0		−2.55	−2.21		−3.45	−2.21		−2.31	−2.21	1.000
0.1		−3.17	−2.45		−3.97	−2.18		−2.94	−2.51	0.773
0.2		−3.79	−2.56		−4.41	−1.97		−3.57	−2.71	0.650
0.3		−4.26	−2.55		−4.64	−1.63		−4.11	−2.81	0.597
0.4		−4.52	−2.49		−4.63	−1.26		−4.45	−2.85	0.595
0.5		−4.61	−2.39		−4.45	−0.93		−4.62	−2.86	0.627
0.6		−4.59	−2.30		−4.20	−0.66		−4.69	−2.85	0.682
0.7		−4.53	−2.19		−3.95	−0.41		−4.72	−2.84	0.751
0.8		−4.46	−2.15		−3.70	−0.28		−4.74	−2.88	0.830
0.9		−4.39	−2.13		−3.49	−0.15		−4.75	−2.94	0.914
1.0		−4.31	−2.10		−3.29	−0.03		−4.76	−2.99	1.000
(f) n-Heptane (1) + Benzene (2) (T = 298.15 K; S = a,4; I = d,4; B = 4,5)										
0	−1.58	−2.73	−2.21	−3.24	−3.58	−2.21	−0.37	−2.13	−2.21	1.000
0.1	−1.61	−3.09	−2.35	−3.16	−3.73	−2.09	−0.37	−2.54	−2.50	0.805
0.2	−1.85	−3.34	−2.40	−3.30	−3.77	−1.90	−0.57	−2.90	−2.74	0.715
0.3	−2.53	−3.46	−2.37	−3.65	−3.67	−1.67	−1.41	−3.17	−2.92	0.690
0.4	−2.85	−3.48	−2.29	−3.74	−3.50	−1.43	−1.88	−3.36	−3.03	0.702
0.5	−3.05	−3.44	−2.18	−3.73	−3.28	−1.20	−2.24	−3.49	−3.10	0.737
0.6	−3.15	−3.36	−2.06	−3.64	−3.07	−0.98	−2.50	−3.58	−3.15	0.784
0.7	−3.17	−3.27	−1.94	−3.51	−2.87	−0.80	−2.70	−3.65	−3.19	0.837
0.8	−3.15	−3.17	−1.87	−3.35	−2.69	−0.71	−2.84	−3.72	−3.28	0.892
0.9	−3.09	−3.09	−1.82	−3.19	−2.53	−0.61	−2.94	−3.79	−3.38	0.947
1.0	−3.01	−3.01	−1.78	−3.01	−2.40	−0.53	−3.01	−3.86	−3.48	1.000

Table 1 (Continued)

	mass-fixed			volume-fixed			number-fixed			
x_1	$D^d_{11}/(10^{-9}\ m^2 \cdot s^{-1})$	$D^d_{12}/(10^{-9}\ m^2 \cdot s^{-1})$	$D^d_{22}/(10^{-9}\ m^2 \cdot s^{-1})$	$D^d_{11}/(10^{-9}\ m^2 \cdot s^{-1})$	$D^d_{12}/(10^{-9}\ m^2 \cdot s^{-1})$	$D^d_{22}/(10^{-9}\ m^2 \cdot s^{-1})$	$D^d_{11}/(10^{-9}\ m^2 \cdot s^{-1})$	$D^d_{12}/(10^{-9}\ m^2 \cdot s^{-1})$	$D^d_{22}/(10^{-9}\ m^2 \cdot s^{-1})$	$[\partial \ln a_i/\partial \ln x_i]_{T,P}$
(g) Octamethylcyclotetrasiloxane (1) + Carbon Tetrachloride (2) (T = 298.15 K; S = a,b,12; I = d,10; B = 11)										
0	−1.34	−1.39	−1.30	−3.17	−2.31	−1.30	0	−0.72	−1.30	1.000
0.1	−1.15	−1.20	−1.10	−2.25	−1.60	−0.79	−0.07	−0.74	−1.28	1.027
0.2	−1.00	−1.01	−0.96	−1.68	−1.14	−0.54	−0.15	−0.74	−1.27	1.082
0.3	−0.85	−0.88	−0.84	−1.29	−0.87	−0.38	−0.18	−0.75	−1.25	1.107
0.4	−0.73	−0.78	−0.74	−1.02	−0.69	−0.26	−0.20	−0.76	−1.23	1.115
0.5	−0.65	−0.69	−0.66	−0.83	−0.55	−0.20	−0.24	−0.76	−1.22	1.125
0.6	−0.57	−0.62	−0.56	−0.69	−0.46	−0.12	−0.26	−0.78	−1.19	1.101
0.7	−0.51	−0.57	−0.47	−0.58	−0.39	−0.06	−0.29	−0.80	−1.16	1.076
0.8	−0.46	−0.51	−0.40	−0.50	−0.34	−0.01	−0.32	−0.81	−1.12	1.050
0.9	−0.41	−0.46	−0.33	−0.43	−0.29	0.04	−0.35	−0.81	−1.08	1.025
1.0	−0.37	−0.42	−0.26	−0.37	−0.25	0.08	−0.37	−0.80	−1.04	1.000
(h) Acetone (1) + Chloroform (2) (T = 298.15 K; S = c,13; I = d,14; B = 15)										
0	−2.61	−1.14	−2.44	−4.52	−2.10	−2.44	−5.0	−2.35	−2.44	1.000
0.1	−1.94	−1.14	−2.63	−3.83	−1.95	−2.49	−4.20	−2.11	−2.45	1.265
0.2		−1.39	−2.91		−2.18	−2.58		−2.30	−2.51	1.286
0.3		−1.58	−3.26		−2.25	−2.70		−2.33	−2.59	1.374
0.4	−1.73	−1.73	−3.71	−3.48	−2.21	−2.91	−3.70	−2.25	−2.77	1.491
0.5	−1.91	−1.99	−4.28	−3.55	−2.25	−3.17	−3.74	−2.26	−2.99	1.524
0.6	−1.88	−2.47	−5.0	−3.48	−2.46	−3.41	−3.64	−2.43	−3.17	1.433
0.7	−1.95	−3.22	−5.9	−3.48	−2.81	−3.54	−3.62	−2.72	−3.22	1.293
0.8	−2.44	−4.11	−7.3	−3.71	−3.08	−3.97	−3.82	−2.93	−3.56	1.215
0.9	−3.32	−5.2	−10	−4.11	−3.32	−5.1	−4.16	−3.10	−4.61	1.162
1.0	−4.52	−7.4	−14	−4.52	−3.95	−6.6	−4.52	−3.62	−5.9	1.000
(i) Methanol (1) + Water (2) (T = 278.15 K; S = a,16; I = a,16; B = 23)										
0	−2.01	−1.56	−1.31	−2.41	−1.75	−1.31	−0.65	−0.88	−1.31	1.000
0.1	−1.23	−0.99	−0.75	−1.46	−1.08	−0.70	−0.42	−0.65	−0.87	1.034
0.2	−1.01	−0.80	−0.51	−1.18	−0.85	−0.43	−0.40	−0.60	−0.71	0.934
0.3	−0.88	−0.79	−0.35	−1.04	−0.80	−0.22	−0.34	−0.67	−0.66	0.777
0.4	−0.88	−0.81	−0.20	−1.03	−0.78	−0.00	−0.39	−0.78	−0.63	0.686
0.5	−0.98	−0.80	−0.09	−1.11	−0.73	0.17	−0.56	−0.86	−0.64	0.692
0.6	−1.10	−0.76	−0.04	−1.20	−0.65	0.28	−0.77	−0.92	−0.69	0.765
0.7	−1.21	−0.73	−0.07	−1.29	−0.59	0.29	−0.97	−0.98	−0.81	0.868
0.8	−1.33	−0.72		−1.38	−0.54		−1.17	−1.06		0.962
0.9	−1.48	−0.75		−1.51	−0.53		−1.39	−1.22		0.999
1.0	−1.67	−0.82	0.25	−1.67	−0.55	0.80	−1.67	−1.46	−1.03	1.000
(j) Methanol (1) + Water (2) (T = 298.15 K; S = a,16; I = a,16; B = 23)										
0	−2.36	−2.78	−2.30	−3.20	−3.20	−2.30	0.07	−1.56	−2.30	1.000
0.1	−1.80	−2.03	−1.45	−2.34	−2.24	−1.33	−0.14	−1.33	−1.70	0.937
0.2	−1.53	−1.72	−0.97	−1.95	−1.83	−0.77	−0.23	−1.29	−1.41	0.838
0.3	−1.44	−1.61	−0.62	−1.79	−1.63	−0.32	−0.34	−1.38	−1.25	0.729
0.4	−1.52	−1.53	−0.33	−1.82	−1.48	0.07	−0.59	−1.48	−1.16	0.665
0.5	−1.67	−1.43	−0.16	−1.91	−1.30	0.33	−0.92	−1.55	−1.16	0.674
0.6	−1.78	−1.31	−0.13	−1.97	−1.12	0.44	−1.21	−1.59	−1.25	0.747
0.7	−1.86	−1.22	−0.16	−2.00	−0.97	0.48	−1.46	−1.64	−1.41	0.850
0.8	−1.99	−1.18	−0.17	−2.07	−0.87	0.54	−1.72	−1.75	−1.58	0.937
0.9	−2.20	−1.20		−2.24	−0.82		−2.06	−1.95		0.979
1.0	−2.37	−1.23	0.70	−2.37	−0.79	1.59	−2.37	−2.19	−1.22	1.000
(k) Acetone (1) + Water (2) (T = 278.15 K; S = a,18; I = a,18; B = 17)										
0	−2.75	−2.35	−1.31	−3.14	−2.55	−1.31	0.49	−0.73	−1.31	1.000
0.1		−1.51	−0.34	−1.81	−1.61	−0.22	0.66	−0.70	−0.80	0.703
0.2	−1.28	−1.64	0.34	−1.60	−1.66	0.63	0.80	−1.06	−0.72	0.404
0.3	−1.54	−1.92	1.36	−1.89	−1.83	1.90	0.93	−1.66	−0.59	0.254
0.4	−1.91	−2.08	2.72	−2.24	−1.85	3.52	0.58	−2.31	−0.22	0.201
0.5	−2.26	−2.37	5.1	−2.56	−1.97	6.2	0.29	−3.28	0.76	0.183
0.6	−2.64	−2.76	9.3	−2.90	−2.14	11	−0.10	−4.67	2.96	0.186
0.7	−3.09	−2.72	13	−3.27	−1.99	15	−1.09	−5.5	5.7	0.233
0.8	−3.44	−2.18	14	−3.53	−1.51	16	−2.30	−5.2		0.356
0.9	−3.57	−1.64	15	−3.61	−1.07	16	−3.12	−4.57		0.582
1.0	−3.50	−1.19	15	−3.50	−0.74	16	−3.50	−3.84	9.5	1.000
(l) Acetone (1) + Water (2) (T = 298.15 K; S = a,12; I = d,14; B = 17)										
0	−2.09	−4.15	−2.29	−3.11	−4.66	−2.29	3.63	−1.29	−2.29	1.000
0.1	−1.43	−2.81	−0.64	−2.23	−3.02	−0.33	3.06	−1.30	−1.64	0.567
0.2	−1.84	−1.93	0.72	−2.55	−1.96	1.35	2.36	−1.25	−1.29	0.319
0.3	−2.44	−1.47	2.20	−3.06	−1.39	3.17	1.63	−1.27	−1.00	0.215
0.4	−2.99	−1.31	3.88	−3.51	−1.15	5.1	0.79	−1.45	−0.59	0.184
0.5	−3.41	−1.33	6.1	−3.82	−1.09	7.7	0.02	−1.84	0.22	0.185
0.6	−3.71	−1.48	9.5	−4.03	−1.13	11	−0.71	−2.51	1.98	0.205
0.7	−4.01	−1.67	12	−4.22	−1.19	13	−1.89	−3.38	3.60	0.276
0.8	−4.24	−1.79	13	−4.35	−1.21	14	−3.00	−4.29	4.66	0.410
0.9	−4.41	−1.80		−4.45	−1.15		−3.87	−5.0		0.639
1.0	−4.52	−1.63	15	−4.52	−0.99	17	−4.52	−5.3	8.1	1.000

Table 1 (Continued)

	mass-fixed			volume-fixed			number-fixed			
x_1	$D_{11}^d/(10^{-9}$ m^{2}s^{-1})	$D_{12}^d/(10^{-9}$ m^{2}s^{-1})	$D_{22}^d/(10^{-9}$ m^{2}s^{-1})	$D_{11}^d/(10^{-9}$ m^{2}s^{-1})	$D_{12}^d/(10^{-9}$ m^{2}s^{-1})	$D_{22}^d/(10^{-9}$ m^{2}s^{-1})	$D_{11}^d/(10^{-9}$ m^{2}s^{-1})	$D_{12}^d/(10^{-9}$ m^{2}s^{-1})	$D_{22}^d/(10^{-9}$ m^{2}s^{-1})	$[\partial \ln a_i/\partial \ln x_i]_{T,P}$
(m) Acetonitrile (1) + Water (2) (T = 278.15 K; S = a,19; I = a,19; B = 20,21)										
0	−8.0	−2.10	−1.31	−8.6	−2.37	−1.31	−5.7	−0.92	−1.31	1.000
0.1		−2.50	−0.68		−2.76	−0.48		−1.40	−1.16	0.391
0.2	4.59	−5.8	1.69	3.00	−6.0	2.70	10	−3.99	−0.59	0.065
0.3	8.6	−13	10	5.5	−13	13	20	−11	2.60	0.011
0.4	11	−22	31	6.4	−21	38	29	−22	12	0.006
0.5	2.84	−15	31	0.64	−13	37	14	−18	14	0.014
0.6	−1.83	−6.7	18	−2.54	−5.6	20	2.31	−9.1	8.8	0.050
0.7	−2.78	−4.71	17	−3.12	−3.84	19	−0.49	−7.4	9.5	0.110
0.8	−2.97	−4.36	24	−3.17	−3.44		−1.49	−7.8		0.187
0.9	−3.18	−4.42	23	−3.27	−3.35		−2.40	−9.0		0.289
1.0	−3.31	−1.90	21	−3.31	−1.36	23	−3.31	−4.32	17	1.000
(n) Acetonitrile (1) + Water (2) (T = 298.15 K; S = a,22; I = a,19; B = 20,21)										
0	−9.9	−3.74	−2.23	−11	−4.39	−2.23	−5.7	−1.65	−2.23	1.000
0.1		−4.70	−1.07		−5.3	−0.62		−2.63	−1.96	0.357
0.2	4.47	−8.1	1.91	2.00	−8.5	3.50	13	−5.6	−1.27	0.092
0.3	11	−18	14	6.0	−18	19	27	−15	3.08	0.027
0.4	0.81	−11	13	−1.48	−10	17	10	−11	3.35	0.038
0.5	−2.44	−8.1	13	−3.69	−7.1	16	3.51	−9.5	4.03	0.060
0.6	−3.11	−8.2	20	−4.04	−6.9	24	1.97	−11	9.2	0.082
0.7	−3.93	−6.7	24	−4.44	−5.4	27	−0.70	−10	13	0.151
0.8	−4.47	−3.93		−4.65	−3.05		−3.14	−7.0		0.368
0.9	−4.45	−3.03	26	−4.52	−2.26	28	−3.92	−6.1	18	0.650
1.0	−4.34	−2.62	27	−4.34	−1.85	28	−4.34	−6.0	20	1.000

uncertain junction of the curves generated by eqs 1 and 2 and the corresponding limiting curves given in our previous paper (*1*).

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