JOURNAL OF CHEMICAL & ENGINEERING DATA

ARTICLE

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Ternary Mutual Diffusion Coefficients of Aqueous $NiCl_2$ + NaCl and $NiCl_2$ + HCl Solutions at 298.15 K

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ABSTRACT: The influence of sodium chloride and hydrochloric acid on the diffusion of aqueous nickel chloride has been investigated by using Taylor dispersion to measure ternary mutual diffusion coefficients for aqueous solutions of $NiCl_2$ + NaCl and $NiCl_2$ + HCl at 298.15 K and carrier concentrations from (0.000 to 0.050) $mol \cdot dm^{-3}$ for each solute. Mutual diffusion coefficients estimated from limiting ionic conductivities using Nernst equations are used to discuss the composition dependence of the diffusion coefficients and the electrostatic mechanism for the strongly coupled diffusion of the solutes.

INTRODUCTION

The diffusion of mixed electrolytes in aqueous solutions and its impact on biological systems is of great interest not only for fundamental purposes but also for many technical fields, such as studies of corrosion in biological systems (e.g., surgical implants and dental alloys[1]), desalination, dissolution, electroplating, and crystallization.[2−5] We have been particularly interested in data on this property for chemical systems involving nickel ions in different mixed electrolytes, such as $NiCl_2$−NaCl−water and $NiCl_2$−HCl−water, for which data are scarce. This work has been motivated by the fact that the nickel ion is one of the most mobile and bioavailable heavy metal ions present in different sources[6−11] (e.g., drinking water, food, active pharmaceutical ingredients and excipients, and dental casting alloys), and by the possibility that the diffusion of nickel salts could produce substantial coupled flows of other dissolved salts. However, to our knowledge, there are only a few publications devoted to the study of binary diffusion in aqueous nickel chloride solutions.[12,13] In the work reported here, Taylor dispersion[14−16] is used to measure ternary mutual diffusion coefficients for aqueous $NiCl_2$ + NaCl and aqueous $NiCl_2$ + HCl solutions at 298.15 K.

EXPERIMENTAL SECTION

Reagents and Solutions. Nickel chloride (Merck > 97 % purity), sodium chloride (Sigma, pro analysi, > 99.5 % purity), and hydrogen chloride (Sociedade Portuense de Drogas, 33.5 wt % HCl) were used without further purification. The solutions for the diffusion measurements were prepared in calibrated volumetric flasks using bidistilled water. The solutions were freshly prepared and deaerated for about 30 min before each set of runs.

Diffusion Measurements. The Taylor dispersion technique is well-described in the literature.[14−22] Only a summary of the experimental procedure is reported here.

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 compositions at the entrance to a Teflon dispersion tube [length 32.799 (± 0.001) m, internal radius r = 0.5570 (± 0.0003) mm]. A flow rate of 0.17 $cm^3 \cdot min^{-1}$ maintained by a metering pump (Gilson model Minipuls 3) gave retention times of about $1.1 \cdot 10^4$ s. The dispersion tube and the injection valve were kept at 298.15 K (± 0.01 K) in an air thermostat. The dispersion of the injected samples was monitored by a differential refractometer (Waters model 2410) at the outlet of the dispersion tube. Detector voltages, $V(t)$, were measured at accurately timed 5 s intervals with a digital voltmeter (Agilent 34401 A) with an IEEE interface.

Mutual diffusion in ternary solutions is described by the equations

$$J_1 = -D_{11}\nabla c_1 - D_{12}\nabla c_2 \tag{1}$$

$$J_2 = -D_{12}\nabla c_1 - D_{22}\nabla c_2 \tag{2}$$

where J_1 and J_2 and ∇c_1 and ∇c_2 are the molar fluxes and the gradients in the concentrations of solute 1 and 2, respectively. Main diffusion coefficients D_{11} and D_{22} give the flux of each solute produced by its own concentration gradient. Cross-diffusion coefficients D_{12} and D_{21} give the coupled flux of each solute driven by a concentration gradient in the other solute. A positive D_{ik} cross-coefficient ($i \neq k$) indicates cocurrent coupled transport of solute i from regions of higher to lower concentrations of solute k. A negative D_{ik} coefficient indicates counter-current coupled transport of solute i from regions of lower to higher concentration of solute k.

Special Issue: Kenneth N. Marsh Festschrift

Received: July 4, 2011
Accepted: September 9, 2011
Published: September 30, 2011

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Table 1. Ternary Mutual Diffusion Coefficients of Aqueous $NiCl_2$ (c_1) + NaCl (c_2) Solutions at 298.15 K[a,b]

c_1	c_2	X_1	D_{11}	D_{12}	D_{21}	D_{22}	D_{12}/D_{22}[c]	D_{21}/D_{11}[d]
0.000	0.050	0.00	0.728 ± 0.010	0.050 ± 0.044	0.246 ± 0.010	1.419 ± 0.001	0.035	0.338
0.001	0.050	0.02	0.712 ± 0.017	−0.003 ± 0.003	0.211 ± 0.026	1.491 ± 0.001	−0.002	0.296
0.001	0.010	0.10	0.719 ± 0.008	0.045 ± 0.030	0.243 ± 0.007	1.419 ± 0.022	0.032	0.338
0.001	0.001	0.50	0.760 ± 0.030	0.142 ± 0.070	0.237 ± 0.050	1.461 ± 0.053	0.097	0.312
0.005	0.005	0.50	0.883 ± 0.002	0.143 ± 0.017	0.190 ± 0.018	1.416 ± 0.002	0.101	0.215
0.010	0.010	0.50	0.870 ± 0.014	0.137 ± 0.050	0.141 ± 0.011	1.334 ± 0.032	0.103	0.162
0.010	0.001	0.90	1.063 ± 0.020	0.320 ± 0.090	0.004 ± 0.001	1.310 ± 0.033	0.244	0.004
0.050	0.000	1.00	1.061 ± 0.005	0.294 ± 0.018	0.001 ± 0.002	1.300 ± 0.015	0.226	0.001

[a] c_1 and c_2 in units of $mol \cdot dm^{-3}$. [b] $D_{ik} \pm S_D$ in units of $10^{-9}\ m^2 \cdot s^{-1}$. [c] D_{12}/D_{22} gives the number of moles of $NiCl_2$ transported per mole of NaCl. [d] D_{21}/D_{11} gives the number of moles of NaCl transported per mole of $NiCl_2$.

Table 2. Ternary Mutual Diffusion Coefficients of Aqueous $NiCl_2$ (c_1) + HCl (c_2) Solutions at 298.15 K[a,b]

c_1	c_2	X_1	D_{11}	D_{12}	D_{21}	D_{22}	D_{12}/D_{22}[c]	D_{21}/D_{11}[d]
0.000	0.050	0.00	0.769 ± 0.016	0.030 ± 0.029	0.422 ± 0.013	2.970 ± 0.031	0.010	0.549
0.001	0.010	0.10	0.734 ± 0.022	−0.226 ± 0.098	0.498 ± 0.018	3.270 ± 0.049	−0.069	0.678
0.010	0.050	0.20	0.739 ± 0.009	−0.363 ± 0.063	0.352 ± 0.010	3.262 ± 0.034	−0.111	0.476
0.010	0.010	0.50	0.863 ± 0.008	−1.212 ± 0.019	0.255 ± 0.004	4.303 ± 0.016	−0.282	0.295
0.050	0.010	0.80	0.872 ± 0.007	−2.538 ± 0.060	0.186 ± 0.008	5.881 ± 0.041	−0.432	0.213
0.050	0.050	0.50	0.825 ± 0.007	−1.364 ± 0.025	0.336 ± 0.007	4.694 ± 0.032	−0.291	0.407
0.050	0.000	1.00	1.056 ± 0.017	−4.107 ± 0.090	0.034 ± 0.032	7.537 ± 0.060	−0.545	0.032

[a] c_1 and c_2 in units of $mol \cdot dm^{-3}$. [b] $D_{ik} \pm S_D$ in units of $10^{-9}\ m^2 \cdot s^{-1}$. [c] D_{12}/D_{22} gives the number of moles of $NiCl_2$ transported per mole of HCl. [d] D_{21}/D_{11} gives the number of moles of HCl transported per mole of $NiCl_2$.

Equation 3[20] was fitted to pairs of dispersion profiles for each carrier solution.

$$V(t) = V_0 + V_1 t + V_{\max}(t_R/t)^{1/2}\left[W_1 \exp\left(-\frac{12D_1(t-t_R)^2}{r^2 t}\right) + (1-W_1)\exp\left(-\frac{12D_2(t-t_R)^2}{r^2 t}\right)\right] \tag{3}$$

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. One of the profiles was generated by injecting solution samples containing a higher concentration of solute 1 relative to the carrier and the other by injecting solution samples containing a higher concentration of solute 2. Ternary mutual D_{ik} coefficients were calculated from the least-squares values of the eigenvalues of the D_{ik} matrix (D_1 and D_2) and the pre-exponential weighting factors (W_1 and $1 - W_1$).

■ RESULTS AND DISCUSSION

Aqueous $NiCl_2$ (1) + NaCl (2) Solutions. Ternary mutual diffusion coefficients were measured six times at each composition and averaged. The results for the aqueous $NiCl_2$ + NaCl solutions and aqueous $NiCl_2$ + HCl solutions are summarized in Tables 1 and 2, respectively. The main diffusion coefficients D_{11} and D_{22} were generally reproducible to within $\pm\ 0.030 \cdot 10^{-9}\ m^2 \cdot s^{-1}$. The cross diffusion coefficients were reproducible to within about $\pm\ 0.050 \cdot 10^{-9}\ m^2 \cdot s^{-1}$.

As indicated in Table 1, D_{11} values are smaller than D_{22} values. In general, D_{11} and D_{22} increase and decrease, respectively, with the solute fraction of $NiCl_2$, defined as $X_1 = c_1/(c_1 + c_2)$. The cross-coefficients are generally positive, indicating cocurrent coupled flows. In the limit $X_1 \to 0$, however, D_{12} is zero because NaCl concentration gradients cannot drive coupled flows of $NiCl_2$ in solutions that do not contain $NiCl_2$. Similarly, D_{21} is zero in the limit $X_1 \to 1$. The values of the ratio D_{12}/D_{22} show that a mole of diffusing NaCl cotransports at most 0.2 mol of $NiCl_2$, whereas the values of D_{21}/D_{11} show that a mole of diffusing $NiCl_2$ can cotransport up to 0.3 mol of NaCl.

Table 3. Limiting Ionic Conductivities[24,25] and Diffusion Coefficients[a] at 298.15 K

	λ_i	D_i
ion	$10^{-4}\ S \cdot m^2 \cdot mol^{-1}$	$10^{-9}\ m^2 \cdot s^{-1}$
Ni^{2+}	53.3	0.71
Na^{+}	50.1	1.33
H^{+}	350.1	9.32
Cl^{-}	76.3	2.03

[a] Calculated from $D_i = RT\lambda_i/z_i F^2$.

Nernst equations[23] 4 to 7 provide values of the D_{ik} coefficients in the limit of infinite dilution. These equations are very useful for qualitatively understanding the main features of the results, including the mechanism of coupled diffusion and the composition dependence of the D_{ik} coefficients.

$$D_{11}^{o} = D_{Ni} + t_{Ni}(D_{Cl} - D_{Ni}) \tag{4}$$

$$D_{21}^{o} = 2t_{Na}(D_{Cl} - D_{Ni}) \tag{5}$$

$$D_{12}^{o} = t_{Ni}(D_{Cl} - D_{Na})/2 \tag{6}$$

$$D_{22}^{o} = D_{Na} + t_{Na}(D_{Cl} - D_{Na}) \tag{7}$$

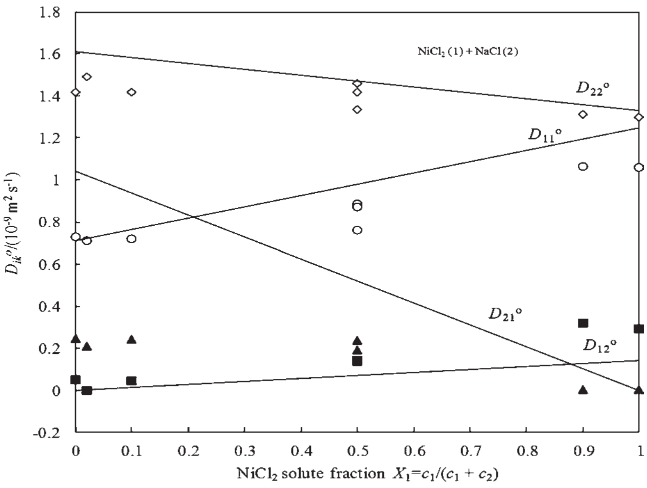

Figure 1. Ternary mutual diffusion coefficients of aqueous $NiCl_2$ (c_1) + NaCl (c_2) solutions at 298.15 K plotted against the solute fraction of $NiCl_2$. Calculated limiting $D°_{ik}$ values (eqs 4 to 7), solid line. Measured values: ○, D_{11}; ■, D_{12}; ▲, D_{21}; ◇, D_{22}.

$$t_{Ni} = \frac{4c_1 D_{Ni}}{4c_1 D_{Ni} + c_2 D_{Na} + (2c_1 + c_2) D_{Cl}} \tag{8}$$

$$t_{Na} = \frac{c_2 D_{Na}}{4c_1 D_{Ni} + c_2 D_{Na} + (2c_1 + c_2) D_{Cl}} \tag{9}$$

D_{Ni}, D_{Na}, and D_{Cl} are the limiting diffusion coefficients of the Ni^{2+}, Na^+, and Cl^- ions, respectively, and t_{Ni} and t_{Na} represent the fraction of the total current carried by the Ni^{2+} and Na^+ ions, respectively. Conveniently, the ionic diffusion coefficients used in the Nernst equations can be evaluated from limiting ionic conductivities (see Table 3).

Each main $D°_{ii}$ coefficient is the sum of a pure diffusion contribution and an electrostatic contribution from the drift of ions in the electric field (diffusion potential gradient) generated by mutual diffusion.[20,23] $D°_{11}$, for example, is the sum of the pure-diffusion term D_{Ni} for the Ni^{2+} ions and the electrostatic term $t_{Ni}(D_{Cl} - D_{Ni})$ for the migration of Ni^{2+} ions in the electric field produced by $NiCl_2$ concentration gradients. Cross-diffusion coefficients $D°_{12}$ and $D°_{21}$, in contrast, are purely electrostatic.

In Figure 1 the limiting Nernst $D°_{ik}$ coefficients and the measured D_{ik} coefficients are plotted against the solute fraction of $NiCl_2$. The Nernst $D°_{11}$ values for composition limits $X_1 = 0$ and $X_1 = 1$ are D_{Ni} (the tracer diffusion coefficient of Ni^{2+} ions in supporting NaCl solutions) and $3D_{Ni}D_{Cl}/(2D_{Na} + D_{Cl})$ (the binary mutual diffusion coefficient of aqueous $NiCl_2$), respectively. Similarly, the limiting $D°_{22}$ values for $X_1 = 0$ and $X_1 = 1$ are $2D_{Na}D_{Cl}/(D_{Na} + D_{Cl})$ and D_{Na}. As the solute fraction of $NiCl_2$ increases from 0 to 1, $D°_{12}$ changes from 0 to $t_{Ni}(D_{Cl} - D_{Na})/2$, and $D°_{21}$ changes from $2t_{Na}(D_{Cl} - D_{Ni})$ to 0.

The diffusion coefficient of the Cl^- ions is larger than that of the Ni^{2+} ions. Consequently, a $NiCl_2$ concentration gradient produces an electric field which slows down the Cl^- ions and speeds up the Ni^{2+} ions to maintain electroneutrality along the diffusion path. If NaCl is present in the solution, the electric field generated by a $NiCl_2$ concentration gradient drives a coupled flow of Na^+ ions in the same direction as the flux of Ni^{2+}, helping to explain the positive values measured for cross-coefficient D_{21}. Similarly, $D_{Cl} > D_{Na}$ and $D_{12} > 0$. But, as shown in Figure 1, only qualitative agreement is obtained for the predicted Nernst $D°_{ik}$ coefficients and the measured D_{ik} coefficients. Nonideal solution behavior (which changes the driving forces for diffusion) and electrophoretic terms are not included in the Nernst limiting $D°_{ik}$ estimates.

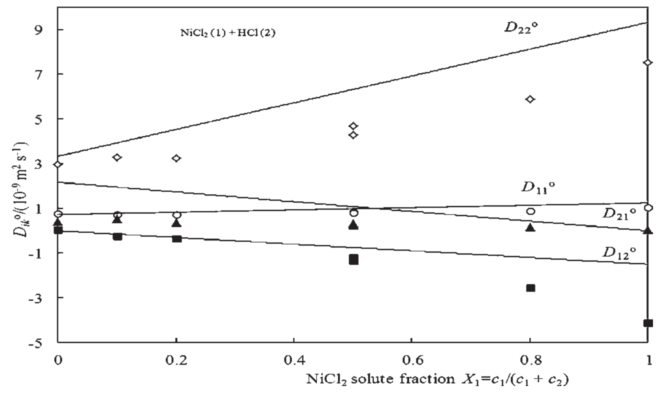

Figure 2. Ternary mutual diffusion coefficients of aqueous $NiCl_2$ (c_1) + HCl (c_2) solutions at 298.15 K plotted against the solute fraction of $NiCl_2$. Calculated limiting $D°_{ik}$ values (eqs 4 and 10 to 12), solid line. Measured values: ○, D_{11}; ■, D_{12}; ▲, D_{21}; ◇, D_{22}.

Aqueous $NiCl_2$ (1) + HCl (2) Solutions. The ternary diffusion coefficients measured for aqueous $NiCl_2$ (c_1) + HCl (c_2) solutions are summarized in Table 2. The D_{22} values are considerably larger than the D_{11} values and increase dramatically with the solute fraction of $NiCl_2$. Also, cross-coefficient D_{12} becomes very large and negative with increasing solute fraction of $NiCl_2$. These results are a consequence of the relatively large mobility of aqueous H^+ ions from the anomalous mechanism for proton transport in water. In the limit $X_1 \rightarrow 1$, for example, D_{22} is the tracer diffusion coefficient of H^+ in $NiCl_2$ solutions. Also, the relatively strong electric field generated by a concentration gradient in HCl, to slow down the H^+ ions drives large counter-current fluxes of Ni^{2+}. However, in the limit $X_1 \rightarrow 0$, D_{12} is zero because HCl concentration gradients cannot drive coupled flows of $NiCl_2$ in solutions that do not contain $NiCl_2$. Similarly, D_{21}, is zero when $X_1 \rightarrow 1$. The values of D_{12}/D_{22} indicate that a mole of diffusing HCl counter-transports up to 0.5 mol of $NiCl_2$. From the D_{21}/D_{11} values, a mole of diffusing $NiCl_2$ cotransports at most 0.7 mol of HCl.

In Figure 2, the D_{ik} coefficients measured for the aqueous $NiCl_2$ (c_1) + HCl (c_2) solutions are plotted against the solute fraction of $NiCl_2$ together with the Nernst $D_{ik}°$ coefficients calculated from the eqs 4 and 10 to 12.

$$D^{\circ}_{12} = t_{Ni}(D_{Cl} - D_H)/2 \tag{10}$$

$$D^{\circ}_{21} = 2t_H(D_{Cl} - D_{Ni}) \tag{11}$$

$$D^{\circ}_{22} = D_H + t_H(D_{Cl} - D_H) \tag{12}$$

In the limit of zero solute fraction of $NiCl_2$ ($X_1 = 0$), $D°_{11}$ is the tracer diffusion coefficient of Ni^{2+} ions in supporting solutions of HCl. In the other limiting case ($X_1 = 1$), $D°_{11}$ is the binary mutual diffusion coefficient of aqueous $NiCl_2$ solutions. Similarly, the Nernst $D°_{22}$ values in these composition limits are D_H and $2D_HD_{Cl}/(D_H + D_{Cl})$, respectively.

CONCLUSIONS

The effects of added sodium chloride and hydrochloric acid on the diffusion of aqueous nickel chloride have been investigated by measuring ternary mutual diffusion coefficients for aqueous solutions of $NiCl_2$ + NaCl and $NiCl_2$ + HCl. The measured D_{ik} coefficients have been compared with limiting D°_{ik} coefficients calculated from limiting ionic conductivities using Nernst equations to help understand the main features of the results, including the mechanism of coupled diffusion and the composition dependence of the D_{ik} coefficients.

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Funding Sources

One of the authors (M.A.E.) is grateful to the University of Alcalá (Spain) for the financial assistance (Mobility Grants for Researchers Program). C.I.A.V.S. is grateful for SFRH/BD/45669/2008 from "Fundação para a Ciência e Tecnologia". D.G.L. acknowledges financial support from the Natural Sciences and Engineering Research Council.

Notes

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