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Activity Coefficients of Hydrochloric Acid in Concentrated Electrolyte Solutions. 2. $HCl + BaCl_2 + KCl + H_2O$, $HCl + LiCl + KCl + H_2O$, and $HCl + NaCl + KCl + H_2O$ at 298.15 K

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Electromotive force have been measured using a Harned cell without a liquid junction of the type $Pt,H_2(0.101\ 325MPa)|HCl(I_A),MeCl(I_B),KCl(I_C)|AgCl,Ag$ with Me = Ba, Li, and Na, respectively, at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1} and at a temperature of 298.15 K. In all the measurements, at each constant total ionic strength, the ionic strength of MeCl (M = Ba, Li, and Na) is taken as 20% of the total ionic strength. From the emf measurements, the mean activity coefficients of hydrochloric acid in quarternary mixtures of $HCl + BaCl_2 + KCl + H_2O$, $HCl + LiCl + KCl + H_2O$, and $HCl + NaCl + KCl + H_2O$ have been determined by the Nernst equation. It is found that the logarithm of the mean activity coefficient of hydrochloric acid in quarternary mixtures studied in this work varies linearly with the ionic strength of KCl, when the total ionic strength of the mixture and the ionic strength of MeCl (Me = Ba, Li, and Na) are constant. An equation is derived which can be used to adequately describe the mean activity coefficients of hydrochloric acid in quarternary mixtures of $HCl + BaCl_2 + KCl + H_2O$, $HCl + LiCl + KCl + H_2O$, and $HCl + NaCl + KCl + H_2O$ at constant total ionic strengths up to 7 mol kg^{-1} and at a temperature of 298.15 K.

Introduction

Knowledge of the mean activity coefficients of hydrochloric acid in mixed aqueous electrolyte solutions in a wide range of concentrations is of fundamental importance for understanding various physicochemical phenomena occurring in hydrometallurgy, electrochemistry, biochemical systems, and other aqueous mixtures of practical and theoretical interest. The electromotive force method has been demonstrated to be precise and useful for determining the mean activity coefficients of hydrochloric acid in aqueous mixtures of hydrochloric acid and metal chloride salts (Harned and Owen, 1958; Robinson and Stokes, 1970; Roy et al., 1983; Pitzer, 1991).

In previous work (Jiang, 1995) on the logarithm of the mean activity coefficient of hydrochloric acid, log γ_A, in concentrated aqueous electrolyte solutions, the values of log γ_A in ternary aqueous solutions of $HCl + NaCl + H_2O$, $HCl + LiCl + H_2O$, and $HCl + BaCl_2 + H_2O$, at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1} and at a temperature of 298.15 K, have been determined from the measurements of electromotive force using a Harned cell without a liquid junction of the type

$$Pt,H_2(0.101\ 325\ MPa)|HCl(I_A),MeCl(I_B)|AgCl,Ag \qquad (A)$$

with Me = Na, Li, and Ba, respectively. Harned's equation has been used in fitting log γ_A, and it has been found that the mean activity coefficients of hydrochloric acid in solutions of $HCl + NaCl + H_2O$, $HCl + LiCl + H_2O$, and $HCl + BaCl_2 + H_2O$ at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1} obey Harned's rule. As a continuation of this study, in the present work, the emf measurements of $HCl + BaCl_2 + KCl + H_2O$, $HCl + LiCl + KCl + H_2O$, and $HCl + NaCl + KCl + H_2O$ are made with a Harned cell without a liquid junction of the type

$$Pt,\ H_2(0.101\ 325\ MPa)|HCl(I_A),MeCl(I_B),KCl(I_C)|AgCl,Ag \qquad (B)$$

with Me = Ba, Li, and Na, respectively, at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1} and at a temperature of 298.15 K, where I_A, I_B, and I_C are the ionic strengths of HCl, MeCl (Me = Ba, Li, and Na), and KCl, respectively. The logarithm of the mean activity coefficient of hydrochloric acid in these quarternary mixtures, at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1}, are calculated from the measured emf data, using the Nernst equation.

Experimental Section

Barium chloride, lithium chloride, sodium chloride, and potassium chloride are reagent grade, purified by recrystallization three times from water, dried, and stored before use. The preparation of the cell solution, electrode construction, emf reading, and other experimental details have been described previously (Jiang, 1995).

The experiments are performed at four constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1} for each mixture. For a given constant total ionic strength, the ionic strength of MeCl (Me = Ba, Li, and Na) are also taken to be constant, with a value of 20% of the constant total ionic strength.

Results and Discussion

Tables 1−3 present the emf measurements of cell B for mixtures of $HCl + BaCl_2 + KCl + H_2O$, $HCl + LiCl + KCl + H_2O$, and $HCl + NaCl + KCl + H_2O$, respectively, at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1} and at a temperature of 298.15 K. The potential data are corrected to a hydrogen gas pressure of 0.101 325 MPa.

The reaction occurring in cell B is

$$(1/2)H_2 + AgCl \rightleftharpoons Ag + H^+ + Cl^- \qquad (1)$$

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Table 1. Experimental Harned Cell Potential, E, and log γ_A of HCl (A) + $BaCl_2$ (B) + KCl (C) + H_2O at Each Constant Total Ionic Strength, I, and at 298.15 K

I_A/(mol kg^{-1})	I_B/(mol kg^{-1})	I_C/(mol kg^{-1})	E/V	log γ_A
$I = 4.0$ mol kg^{-1}				
3.20	0.80	0.00	0.125 90	0.190 79
2.88	0.80	0.32	0.131 21	0.168 74
2.56	0.80	0.64	0.136 91	0.146 16
2.24	0.80	0.96	0.142 74	0.125 83
1.92	0.80	1.28	0.149 23	0.104 51
1.60	0.80	1.60	0.156 27	0.084 50
1.28	0.80	1.92	0.164 42	0.064 12
0.96	0.80	2.24	0.174 20	0.043 86
0.64	0.80	2.56	0.186 93	0.024 33
0.32	0.80	2.88	0.207 01	0.005 06
$I = 5.0$ mol kg^{-1}				
4.00	1.00	0.00	0.100 42	0.309 24
3.60	1.00	0.40	0.106 54	0.280 37
3.20	1.00	0.80	0.113 10	0.250 53
2.80	1.00	1.20	0.119 61	0.224 47
2.40	1.00	1.60	0.126 87	0.196 61
2.00	1.00	2.00	0.134 56	0.171 15
1.60	1.00	2.40	0.143 40	0.144 85
1.20	1.00	2.80	0.153 91	0.118 49
0.80	1.00	3.20	0.167 27	0.093 58
0.40	1.00	3.60	0.187 97	0.069 10
$I = 6.0$ mol kg^{-1}				
4.80	1.20	0.00	0.076 44	0.432 82
4.32	1.20	0.48	0.083 43	0.396 57
3.84	1.20	0.96	0.090 91	0.358 92
3.36	1.20	1.44	0.098 15	0.326 70
2.88	1.20	1.92	0.106 24	0.291 85
2.40	1.20	2.40	0.114 62	0.260 55
1.92	1.20	2.88	0.124 20	0.227 98
1.44	1.20	3.36	0.135 47	0.195 18
0.96	1.20	3.84	0.149 50	0.164 62
0.48	1.20	4.32	0.170 85	0.134 69
$I = 7.0$ mol kg^{-1}				
5.60	1.40	0.00	0.053 42	0.560 42
5.04	1.40	0.56	0.061 36	0.516 24
4.48	1.40	1.12	0.069 82	0.470 25
3.92	1.40	1.68	0.077 85	0.431 41
3.36	1.40	2.24	0.086 81	0.389 13
2.80	1.40	2.80	0.095 93	0.351 62
2.24	1.40	3.36	0.106 30	0.312 42
1.68	1.40	3.92	0.118 36	0.272 87
1.12	1.40	4.48	0.133 10	0.236 38
0.56	1.40	5.04	0.155 11	0.200 77

Table 2. Experimental Harned Cell Potential, E, and log γ_A of HCl (A) + LiCl (B) + KCl (C) + H_2O at Each Constant Total Ionic Strength, I, and at 298.15 K

I_A/(mol kg^{-1})	I_B/(mol kg^{-1})	I_C/(mol kg^{-1})	E/V	log γ_A
$I = 4.0$ mol kg^{-1}				
3.20	0.80	0.00	0.128 37	0.242 93
2.88	0.80	0.32	0.133 72	0.220 61
2.56	0.80	0.64	0.139 48	0.197 46
2.24	0.80	0.96	0.145 31	0.177 22
1.92	0.80	1.28	0.151 84	0.155 47
1.60	0.80	1.60	0.158 88	0.135 56
1.28	0.80	1.92	0.167 05	0.114 91
0.96	0.80	2.24	0.176 90	0.094 14
0.64	0.80	2.56	0.189 64	0.074 44
0.32	0.80	2.88	0.209 74	0.055 01
$I = 5.0$ mol kg^{-1}				
4.00	1.00	0.00	0.101 25	0.375 30
3.60	1.00	0.40	0.107 42	0.345 99
3.20	1.00	0.80	0.114 08	0.315 32
2.80	1.00	1.20	0.120 59	0.289 29
2.40	1.00	1.60	0.127 92	0.260 79
2.00	1.00	2.00	0.135 61	0.235 34
1.60	1.00	2.40	0.144 50	0.208 61
1.20	1.00	2.80	0.155 10	0.181 50
0.80	1.00	3.20	0.168 50	0.156 28
0.40	1.00	3.60	0.189 23	0.131 49
$I = 6.0$ mol kg^{-1}				
4.80	1.20	0.00	0.075 59	0.513 00
4.32	1.20	0.48	0.082 67	0.476 10
3.84	1.20	0.96	0.090 28	0.437 33
3.36	1.20	1.44	0.097 53	0.405 02
2.88	1.20	1.92	0.105 72	0.369 28
2.40	1.20	2.40	0.114 11	0.337 89
1.92	1.20	2.88	0.123 77	0.304 67
1.44	1.20	3.36	0.135 16	0.270 85
0.96	1.20	3.84	0.149 25	0.239 80
0.48	1.20	4.32	0.170 65	0.209 39
$I = 7.0$ mol kg^{-1}				
5.60	1.40	0.00	0.050 89	0.654 90
5.04	1.40	0.56	0.058 93	0.609 83
4.48	1.40	1.12	0.067 56	0.562 39
3.92	1.40	1.68	0.075 62	0.523 31
3.36	1.40	2.24	0.084 72	0.479 87
2.80	1.40	2.80	0.093 87	0.442 11
2.24	1.40	3.36	0.104 34	0.402 01
1.68	1.40	3.92	0.116 56	0.361 14
1.12	1.40	4.48	0.131 38	0.323 94
0.56	1.40	5.04	0.153 48	0.287 62

The cell potential, E, is given by the Nernst equation

$$E = E^0 - \frac{RT}{F} \ln a_H a_{Cl} \tag{2}$$

where a refers to activity. Values of R and F are 8.314 cm^3 MPa mol^{-1} K^{-1} and 96 487 C mol^{-1}, respectively. The value of E^0, the standard potential of the silver−silver chloride electrode on a molality basis, is taken as 0.222 59 V in this study, as reported by Roy et al. (1980, 1992) and Simonson et al. (1987).

The logarithm of the mean activity coefficient of hydrochloric acid, log γ_A, is determined from the cell potential, by rearranging the Nernst equation to give

$$\log \gamma_A = \frac{F(E^0 - E)}{2RT \ln 10} - \frac{1}{2} \log[(I + 2I_B)I_A] \tag{3}$$

for HCl + $BaCl_2$ + KCl + H_2O, and

$$\log \gamma_A = \frac{F(E^0 - E)}{2RT \ln 10} - \frac{1}{2} \log(II_A) \tag{4}$$

for HCl + LiCl + KCl + H_2O and HCl + NaCl + KCl + H_2O, where $I = I_A + I_B + I_C$, the total ionic strength of the mixture.

The values of log γ_A in HCl + $BaCl_2$ + KCl + H_2O, HCl + LiCl + KCl + H_2O, and HCl + NaCl + KCl + H_2O, at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1} and at temperature of 298.15 K, are given in Tables 1−3.

In Figures 1−3, log γ_A values from Tables 1−3 are plotted against the ionic strength of KCl at each constant total ionic strength, for HCl + $BaCl_2$ + KCl + H_2O, HCl + LiCl + KCl + H_2O, and HCl + NaCl + KCl + H_2O, respectively. It can be seen from these figures that log γ_A varies linearly with the ionic strength of KCl in quarternary mixtures studied here when the total ionic strength and the ionic strength of MeCl (Me = Ba, Li, and Na) are taken to be constant:

$$\log \gamma_A = \log \gamma_A^{(3)} - \alpha_{AC} I_C \tag{5}$$

where $\gamma_A^{(3)}$ refers to the mean activity coefficient of hydrochloric acid in ternary solutions of HCl + $BaCl_2$ + H_2O, HCl + LiCl + H_2O, and HCl + NaCl + H_2O, respectively, based on the condition that the constant total ionic strength is equal to the constant total ionic strength of the corresponding quarternary mixture, and the ionic strength of MeCl (Me = Ba, Li, and Na) is also constant and is equal

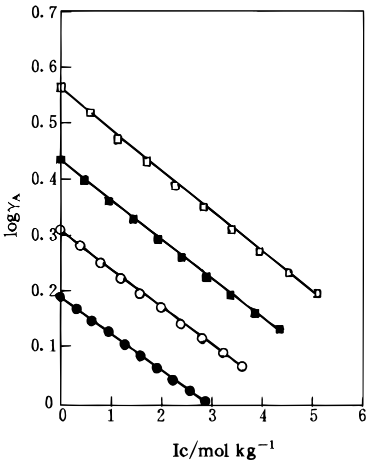

Figure 1. Variation of the logarithm of the mean activity coefficient of hydrochloric acid, log γ_A, with the ionic strength of KCl, I_C, in HCl + $BaCl_2$ + KCl + H_2O, at each constant total ionic strength, I, and ionic strength of $BaCl_2$ at 298.15 K: (●) I = 4 mol kg^{-1}; (○) I = 5 mol kg^{-1}; (■) I = 6 mol kg^{-1}; (□) I = 7 mol kg^{-1}.

Table 3. Experimental Harned Cell Potential, *E*, and log γ_A of HCl (A) + NaCl (B) + KCl (C) + H_2O at Each Constant Total Ionic Strength, *I*, and at 298.15 K

I_A/(mol kg^{-1})	I_B/(mol kg^{-1})	I_C/(mol kg^{-1})	E/V	log γ_A
		I = 4.0 mol kg^{-1}		
3.20	0.80	0.00	0.131 10	0.219 87
2.88	0.80	0.32	0.136 36	0.198 29
2.56	0.80	0.64	0.142 02	0.176 02
2.24	0.80	0.96	0.147 77	0.156 37
1.92	0.80	1.28	0.154 21	0.135 43
1.60	0.80	1.60	0.161 18	0.116 12
1.28	0.80	1.92	0.169 26	0.096 21
0.96	0.80	2.24	0.179 01	0.076 27
0.64	0.80	2.56	0.191 67	0.057 27
0.32	0.80	2.88	0.211 69	0.038 55
		I = 5.0 mol kg^{-1}		
4.00	1.00	0.00	0.104 95	0.344 03
3.60	1.00	0.40	0.110 99	0.315 88
3.20	1.00	0.80	0.117 48	0.286 55
2.80	1.00	1.20	0.123 88	0.261 47
2.40	1.00	1.60	0.131 06	0.234 21
2.00	1.00	2.00	0.138 64	0.209 73
1.60	1.00	2.40	0.147 40	0.184 15
1.20	1.00	2.80	0.157 84	0.158 31
0.80	1.00	3.20	0.171 11	0.134 19
0.40	1.00	3.60	0.191 71	0.110 51
		I = 6.0 mol kg^{-1}		
4.80	1.20	0.00	0.080 39	0.472 48
4.32	1.20	0.48	0.087 26	0.437 25
3.84	1.20	0.96	0.094 65	0.400 37
3.36	1.20	1.44	0.101 74	0.369 46
2.88	1.20	1.92	0.109 71	0.335 49
2.40	1.20	2.40	0.117 94	0.305 52
1.92	1.20	2.88	0.127 41	0.273 96
1.44	1.20	3.36	0.138 58	0.241 94
0.96	1.20	3.84	0.152 48	0.212 48
0.48	1.20	4.32	0.173 70	0.183 66
		I = 7.0 mol kg^{-1}		
5.60	1.40	0.00	0.056 90	0.604 10
5.04	1.40	0.56	0.064 67	0.561 29
4.48	1.40	1.12	0.073 01	0.516 39
3.92	1.40	1.68	0.080 83	0.479 25
3.36	1.40	2.24	0.089 65	0.438 18
2.80	1.40	2.80	0.098 56	0.402 39
2.24	1.40	3.36	0.108 77	0.364 55
1.68	1.40	3.92	0.120 71	0.326 10
1.12	1.40	4.48	0.135 27	0.291 07
0.56	1.40	5.04	0.157 11	0.256 93

to 20% of the constant total ionic strength. The term α_{AC} is a parameter.

The mean activity coefficients of hydrochloric acid presented in Tables 1−3 are used in fitting eq 5 by a least squares method, and log $\gamma_A^{(3)}$ as well as α_{AC} is determined.

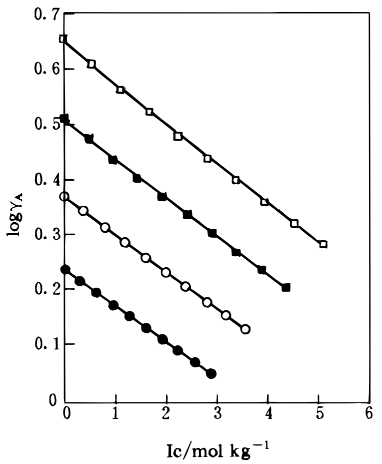

Figure 2. Variation of the logarithm of the mean activity coefficient of hydrochloric acid, log γ_A, with the ionic strength of KCl, I_C, in HCl + LiCl + KCl + H_2O, at each constant total ionic strength, I, and ionic strength of LiCl at 298.15 K: (●) I = 4 mol kg^{-1}; (○) I = 5 mol kg^{-1}; (■) I = 6 mol kg^{-1}; (□) I = 7 mol kg^{-1}.

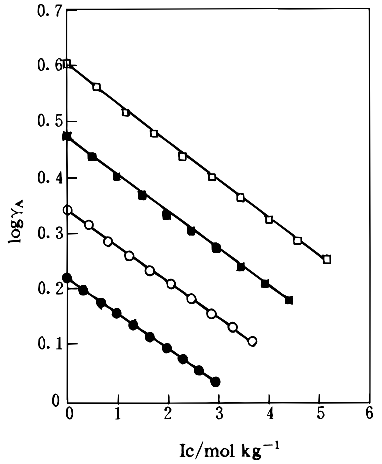

Figure 3. Variation of the logarithm of the mean activity coefficient of hydrochloric acid, log γ_A, with the ionic strength of KCl, I_C, in HCl + NaCl + KCl + H_2O, at each constant total ionic strength, I, and ionic strength of NaCl at 298.15 K: (●) I = 4 mol kg^{-1}; (○) I = 5 mol kg^{-1}; (■) I = 6 mol kg^{-1}; (□) I = 7 mol kg^{-1}.

Table 4. Coefficients log $\gamma_A^{(3)}$ and α_{AC} and the Overall Root Mean Square Deviations σ of HCl (A) + $BaCl_2$ (B) + KCl (C) + H_2O at Each Constant Total Ionic Strength, *I*/(mol kg^{-1}), and at 298.15 K

I	log $\gamma_{Aexp}^{(3)}$ [a]	log $\gamma_{Aexp}^{(3)}$ [b]	log $\gamma_A^{(3)}$	α_{AC}	$10^4\sigma$
4.0	0.189 26	0.190 79	0.19055 ± 0.0003	0.06920 ± 0.0005	3.79
5.0	0.306 77	0.309 24	0.30884 ± 0.0005	0.07253 ± 0.0007	6.82
6.0	0.429 37	0.432 82	0.43224 ± 0.0008	0.07593 ± 0.0009	9.98
7.0	0.555 95	0.560 42	0.55966 ± 0.0010	0.07939 ± 0.0010	13.23

[a] Jiang (1995). [b] This study.

Table 5. Coefficients log $\gamma_A^{(3)}$ and α_{AC} and the Overall Root Mean Square Deviations σ of HCl (A) + LiCl (B) + KCl (C) + H_2O at Each Constant Total Ionic Strength, *I*/(mol kg^{-1}), at 298.15 K

I	log $\gamma_{Aexp}^{(3)}$ [a]	log $\gamma_{Aexp}^{(3)}$ [b]	log $\gamma_A^{(3)}$	α_{AC}	$10^4\sigma$
4.0	0.240 99	0.242 93	0.24261 ± 0.0004	0.06997 ± 0.0007	5.38
5.0	0.372 31	0.375 30	0.37497 ± 0.0007	0.07355 ± 0.0009	8.86
6.0	0.508 90	0.513 00	0.51299 ± 0.0010	0.07721 ± 0.0011	12.46
7.0	0.649 67	0.654 90	0.65399 ± 0.0013	0.08090 ± 0.0012	16.15

[a] Jiang (1995). [b] This study.

In Tables 4−6, log $\gamma_A^{(3)}$ and α_{AC}, together with their standard deviations, and the overall root mean square deviation, σ, of the fit of log γ_A are shown. The experi-

Table 6. Coefficients log $\gamma_A^{(3)}$ and α_{AC} and the Overall Root Mean Square Deviations σ of HCl (A) + NaCl (B) + KCl (C) + H_2O at Each Constant Total Ionic Strength, I/(mol kg^{-1}), at 298.15 K

I	log $\gamma_{Aexp}^{(3)}$ [a]	log $\gamma_{Aexp}^{(3)}$ [b]	log $\gamma_A^{(3)}$	α_{AC}	$10^4\sigma$
4.0	0.218 11	0.219 87	0.21958 ± 0.0004	0.06766 ± 0.0006	4.73
5.0	0.341 29	0.344 03	0.34357 ± 0.0006	0.07068 ± 0.0008	7.99
6.0	0.468 71	0.472 48	0.47183 ± 0.0009	0.07377 ± 0.0010	11.35
7.0	0.599 28	0.604 10	0.60326 ± 0.0012	0.07690 ± 0.0011	14.78

[a] Jiang (1995). [b] This study.

mental log $\gamma_A^{(3)}$ values obtained in this study and in previous work (Jiang, 1995) are also shown in Tables 4−6. It is clear that, at all constant total ionic strengths up to 7.0 mol kg^{-1}, the agreement is acceptable among the correlated and experimental log $\gamma_A^{(3)}$ values, obtained both in the present work and in previous work.

Finally, since the mean activity coefficients of hydrochloric acid in ternary solutions of HCl + NaCl + H_2O, HCl + LiCl + H_2O, and HCl + $BaCl_2$ + H_2O, at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1}, follow Harned's rule (Jiang, 1995), log $\gamma_A^{(3)}$ can be expressed as

$$\log \gamma_A^{(3)} = \log {\gamma_A}^0 - \alpha_{AB} I_B \tag{6}$$

Substituting eq 6 into eq 5 leads to

$$\log \gamma_A = \log {\gamma_A}^0 - \alpha_{AB} I_B - \alpha_{AC} I_C \tag{7}$$

Equation 7 can be used to adequately describe the mean activity coefficients of hydrochloric acid in quarternary mixtures studied in this work at constant total ionic strengths of (4, 5, 6, and 7) mol kg^{-1} and at a temperature of 298.15 K.

Literature Cited

Harned, H. S.; Owen, B. B. *The Physical Chemistry of Electrolyte Solutions*, 3rd ed.; Reinhold: New York, 1958.

Jiang, C. F. Activity Coefficients of Hydrochloric Acid in Concentrated Electrolyte Solutions. 1. HCl + NaCl + H_2O, HCl + LiCl + H_2O, and HCl + $BaCl_2$ + H_2O at 298.15 K. *J. Chem. Eng. Data* **1995**, previous paper in this issue.

Pitzer, K. S. In *Activity Coefficients in Electrolyte Solutions*, 2nd ed.; Pitzer, K. S., Ed.; CRC Press: Boca Raton, FL, 1991.

Robinson, R. A.; Stokes, R. H. *Electrolyte Solutions*, revised ed.; Butterworths: London, 1970.

Roy, R. N.; Gibbons, J. J.; Bliss, D. P., Jr.; Casebolt, R. G.; Baker, B. K. Activity Coefficients for Ternary Systems. VI. The System HCl + $MgCl_2$ + H_2O at Different Temperature; Application of Pitzer's Equation. *J. Solution Chem.* **1980**, *9*, 911−930.

Roy, R. N.; Gibbons, J. J.; Peiper, J. C.; Pitzer, K. S. Thermodynamics of Unsymmetrical Mixed Electrolyte HCl + $LaCl_3$. *J. Phys. Chem.* **1983**, *87*, 2365−2369.

Roy, R. N.; Vogel, K. M.; Good, C. E.; Davis, W. B.; Roy, L. N.; Johnson, D. A.; Felmy, A. R.; Pitzer, K. S. Activity Coefficients in Electrolyte Mixtures: HCl + $ThCl_4$ + H_2O for 5−55 °C. *J. Phys. Chem.* **1992**, *96*, 11065−11072.

Simonson, J. M.; Roy, R. N.; Roy, L. N.; Johnson, D. A. The Thermodynamics of Aqueous Borate Solutions. I. Mixtures of Boric Acid with Sodium or Potassium Borate and Chloride. *J. Solution Chem.* **1987**, *16*, 791−803.

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