SUPPORTING INFORMATION

GIBBS FREE ENERGY-BASED OBJECTIVE FUNCTION FOR ELECTROLYTE ACTIVITY COEFFICIENT MODELS

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Appendix 1. The Activity Coefficient Models

A1.1 Bromley Model

The individual activity coefficient is calculated as:

$$log\gamma_i = -\frac{Az_i^2\sqrt{I}}{1+\sqrt{I}} + F_i \quad \text{(A1)}$$

$$F_i = \sum_j \dot{B}_{ij} Z_{ij} m_j \quad (j\ refers\ to\ the\ ions\ having\ opposite\ charge\ of\ i) \quad \text{(A2)}$$

$$Z_{ij} = \frac{z_i + z_j}{2} \quad \text{(A3)}$$

$$\dot{B}_{ij} = \frac{0.06 + 0.6B_{ij}}{\left(1 + \frac{1.5}{|z_i z_j|} I\right)^2} + B_{ij} \quad \text{(A4)}$$

$$I = \frac{1}{2}\sum_i^{ions} m_i z_i^2 \quad \text{(A5)}$$

where z_i represents the number of charge of i, B_{ij} represents the interaction parameter between opposite-charged ions, I represents the ionic strength and A is Debye-Hückel constant. A and B_{ij} are temperature-dependent parameters. Equation A6 gives the temperature dependence of B_{ij}.

$$B_{ij} = B^* \ln\left(\frac{T-243}{T}\right) + \frac{B_1}{T} + B_2 + B_3 lnT \quad \text{(A6)}$$

Equation A7 gives the temperature dependence of A in natural logarithm scale. Then, it is converted to logarithm of 10 base by equation A8.

$$A_{ln} = 1.131 + 1.335 * 10^{-3} * (T - 273.15) + 1.164 * 10^{-5} * (T - 273.15)^2 \quad \text{(A7)}$$

$$A = \log(\exp(A_{ln})) \quad \text{(A8)}$$

The mean molal activity coefficient is calculated based on individual coefficients as:

$$log\gamma_m = \frac{\sum_i^{ions} m_i log\gamma_i}{\sum_i^{ions} m_i} \quad \text{(A9)}$$

A1.2 Pitzer Model

The osmotic coefficient and mean molal activity coefficient are calculated as:

$$\varphi - 1 = |z_C z_A| f^{\varphi} + m\left(\frac{2v_C v_A}{v_C+v_A}\right) B_{CA}^{\varphi} + m^2 \frac{2(v_C v_A)^{3/2}}{v_C+v_A} C_{CA}^{\varphi} \quad \text{(A10)}$$

$$ln\gamma_{\pm}^{m} = |z_C z_A| f^{\gamma} + \left(\frac{2v_C v_A}{v_C+v_A}\right) B_{CA}^{\gamma} + m^2 \frac{2(v_C v_A)^{3/2}}{v_C+v_A} C_{CA}^{\gamma} \quad \text{(A11)}$$

$$f^{\gamma} = -A_{\varphi}\left[\frac{I^{1/2}}{1+bI^{1/2}} + \frac{2}{b}\ln(1+bI^{1/2})\right] \quad \text{(A12)}$$

$$f^{\varphi} = -A_{\varphi}\frac{I^{1/2}}{1+bI^{1/2}} \quad \text{(A13)}$$

$$B_{CA}^{\gamma} = 2\beta_{CA}^{0} + \frac{2\beta_{CA}^{1}}{\alpha^2 I}\left[1 - e^{-\alpha I^{1/2}}\left(1 + \alpha I^{1/2} - 0.5\alpha^2 I\right)\right] \quad \text{(A14)}$$

$$B_{CA}^{\varphi} = \beta_{CA}^{0} + \beta_{CA}^{1} e^{-\alpha I^{1/2}} \quad \text{(A15)}$$

$$C_{CA}^{\gamma} = 1.5 C_{CA}^{\varphi} \quad \text{(A16)}$$

where β_{CA}^{0}, β_{CA}^{1} and C_{CA}^{φ} are the interaction parameters fitted to the data. α and b have the values of 2 and 1.2, respectively. The temperature dependence of these parameters are expressed as:

$$\beta \; or \; C = p_6 T^6 + p_5 T^5 + p_4 T^4 + p_3 T^3 + p_2 T^2 + p_1 T^1 + p_0 \quad \text{(A17)}$$

Appendix 2. The Model Parameters Determined Through Parameter Fitting

A2.1 Bromley Model

Table A1. The parameters of Bromley model

Salt in water as binary system	The proposed approach	The common approach
NaCl	$B^* = 0.03969$ $B_1 = 2.7225$ $B_2 = 0.6350$ $B_3 = -0.09108$	$B^* = 0.04073$ $B_1 = 2.6586$ $B_2 = 0.6021$ $B_3 = -0.08503$
KBr	$B^* = 0.05241$ $B_1 = 6.4688 \times 10^{-5}$ $B_2 = 0.3648$ $B_3 = -0.04344$	$B^* = 0.05274$ $B_1 = 4.1543$ $B_2 = 0.2404$ $B_3 = -0.02389$
KCl	$B^* = 0.03269$ $B_1 = 0.0004599$ $B_2 = 0.2419$ $B_3 = -0.02857$	$B^* = 0.03056$ $B_1 = 8.0462 \times 10^{-5}$ $B_2 = 0.1427$ $B_3 = -0.01180$

A2.2 Pitzer Model

Table A2. The parameters of Pitzer with the common approach

Salt in water as binary system	β^0_{CA}	β^1_{CA}	C^{φ}_{CA}	
NaCl	6.2188E-15	-3.8303E-13	-1.057E-15	p_6
	-1.0394E-11	7.7115E-10	1.8096E-12	p_5
	6.7051E-09	-6.4616E-07	-1.219E-09	p_4
	-1.9805E-06	0.0002885	3.9703E-07	p_3
	0.0002047	-0.07240	-5.821E-05	p_2
	0.01868	9.6867	0.001455	p_1
	-4.0634	-539.76	0.3338	p_0
KBr	0	0	0	p_6
	-1.7731E-12	0	-1.5152E-11	p_5
	2.7270E-09	-1.2537E-09	2.583E-08	p_4
	-1.5794E-06	1.4618E-06	-1.7598E-05	p_3
	0.0004100	-0.00060400	0.005991	p_2
	-0.04103	0.1027	-1.0191	p_1
	0.3311	-5.5143	69.32	p_0
KCl	-8.2769E-12	0	0	p_6
	1.5840E-08	-1.987E-09	-1.6344E-11	p_5
	-1.2615E-05	3.1974E-06	2.6570E-08	p_4
	0.005352	-0.002055	-1.7252E-05	p_3
	-1.2757	0.6594	0.005593	p_2
	161.97	-105.63	-0.9051	p_1
	-8558.5	6758.5	58.50	p_0

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Salt in water as binary system	β^0_{CA}	β^1_{CA}	C^{φ}_{CA}	
NaCl	-1.6320E-14	-2.5784E-13	3.5505E-15	p_6
	3.3379E-11	5.2789E-10	-7.1441E-12	p_5
	-2.8675E-08	-4.4950E-07	6.0231E-09	p_4
	1.3255E-05	0.0002038	-2.7247E-06	p_3
	-0.003482	-0.05192	0.0006981	p_2
	0.4942	7.0473	-0.09624	p_1
	-29.61	-398.19	5.5906	p_0
KBr	0	0	0	p_6
	0	0	0	p_5
	-4.3921E-09	0	3.4807E-10	p_4
	6.0219E-06	-2.0493E-07	-4.6482E-07	p_3
	-0.003097	0.0002223	0.0002337	p_2
	0.7082	-0.07833	-0.05248	p_1
	-60.71	9.2596	4.4406	p_0
KCl	0	9.9799E-12	0	p_6
	-1.2429E-10	-2.0257E-08	2.224E-11	p_5
	1.9968E-07	1.7082E-05	-3.5218E-08	p_4
	-0.0001281	-0.007661	2.2263E-05	p_3
	0.04099	1.9274	-0.007023	p_2
	-6.5456	-257.92	1.1056	p_1
	417.23	14343.32	-69.49	p_0

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