

Prediction of solid–liquid equilibria in mixed electrolyte aqueous solution by the modified mean spherical approximation

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Abstract

Solid–liquid equilibria in mixed electrolyte aqueous solution have been investigated using available thermodynamic data for solids and for aqueous electrolyte solutions. The mean spherical approximation (MSA) modified by Lu et al. [Fluid Phase Equilib. 85 (1993) 81] is used to calculate the mean ionic activity coefficients in mixed electrolyte solutions at saturation conditions. Solid–liquid equilibria of seven mixed electrolyte systems at 298.15 K are successfully predicted using the modified MSA method with the parameters obtained from activity coefficient data of corresponding single electrolyte solutions. The total average absolute deviation between predicted and experimental values is 5.58%. Furthermore, the predicted results of solid–liquid equilibria for four mixed electrolyte solutions over a range of temperature indicate that the modified MSA method can fairly be used to predict solid–liquid equilibria for mixed electrolyte aqueous solutions at various temperatures.

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1. Introduction

Solid–liquid equilibria of mixed electrolyte aqueous solution play an important role in separation operation for many industrial processes involving electrolyte solutions. To meet engineering requirements, an accurate estimation method is necessary for engineering design because solid–liquid equilibrium data for mixed electrolyte solutions are scarcely available in literature.

To perform the calculation of solid–liquid equilibria for mixed electrolyte aqueous solution, a key problem is how to accurately express the activity coefficient of electrolyte in a saturated solution. In recent years, Several models for activity coefficient of electrolyte solution have been proposed [1–12]. Pitzer model [1] is widely used because it has high accuracy at concentration below 6 mol kg⁻¹, and

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parameters for many single electrolyte solutions have been given in literature. However, it is difficult to use this model to predict thermodynamic properties at various temperatures due to the complex relationship between the parameters and temperature. Chen model [3] and Clegg–Pitzer model [4] can be used to correlate the solid–liquid equilibria in electrolyte solution with high accuracy [3–5]. The drawback of these semi-empirical models [1–4] is that some additional mixing parameters, which must be determined from the experimental data of mixed electrolyte solutions, are required.

Much attention has been paid to statistical mechanic methods, especially for mean spherical approximation (MSA) method because it gives analytical expressions for thermodynamic properties of electrolyte solution. For example, Blum and Høye [6,7] reported an expression for excess free energy and activity coefficients for ionic mixtures with concentration less than 2 mol kg⁻¹; Renon and co-workers [8,9] applied the MSA with a non-primitive model to calculate osmotic coefficients of single and mixed electrolyte solutions; Gao et al. [10] combined non-primitive MSA with perturbation theory to calculate activity coefficient of electrolyte and solubility of gas in aqueous electrolyte solutions; Taghikhani and Vera [11] used a Kelvin hard sphere-mean spherical approximation model to correlate activity coefficient for aqueous electrolyte solutions. In their study, the maximum concentration of electrolyte solution is 6 mol kg⁻¹. This is not adequate for the calculation of solid–liquid equilibria. In order to apply MSA to electrolyte solutions at high concentrations (up to saturation), Lu et al. [12] improved MSA by introducing an effective diameter of cation. The modified MSA has been used to calculate activity coefficient and surface tension of single and mixed electrolyte solutions at very high concentration [12,13]. In this work, the modified MSA is used to investigate solid–liquid equilibria of mixed electrolyte solutions without any mixing parameters.

2. Theory

2.1. Solid–liquid equilibrium in aqueous electrolyte solution

According to the principle of thermodynamics, solid–liquid equilibrium at a given temperature and pressure may be described by equality of chemical potential of individual components in solid and liquid phase

$$\mu_S = \mu_L \quad (1)$$

Considering the dissociation equilibrium



the criterion for equilibrium is

$$\mu_L - (\nu_M \mu_M + \nu_A \mu_A + \nu_0 \mu_{H_2O}) = 0 \quad (2)$$

where

$$\mu_i = \mu_i^0 + RT \ln a_i \quad (3)$$

are the chemical potential of species *i*. The equilibrium constant of the dissociation is obtained from Eqs. (2) and (3)

$$\ln K = -\frac{\nu_M \mu_M^0 + \nu_A \mu_A^0 + \nu_0 \mu_{H_2O}^0}{RT} + \frac{\mu_L^0}{RT} = \nu_M \ln a_M + \nu_A \ln a_A + \nu_0 \ln a_{H_2O} - \ln a_L \quad (4)$$

If the stable solid hydrate $M_{\nu_M}A_{\nu_A} \cdot \nu_0H_2O$ is taken as standard state, then

$$\mu_S = \mu_S^0 \quad (5)$$

$$\mu_L = \mu_S^0 + RT \ln a_L \quad (6)$$

Incorporating Eqs. (1), (5) and (6), we can obtain the activity of liquid $M_{\nu_M}A_{\nu_A} \cdot \nu_0H_2O$

$$\ln a_L = 0 \quad (7)$$

Therefore, Eq. (4) can be expressed as

$$\ln K = \nu_M \ln a_M + \nu_A \ln a_A + \nu_0 \ln a_{H_2O} = \nu_M \ln(m_M \gamma_M) + \nu_A \ln(m_A \gamma_A) + \nu_0 \ln a_{H_2O} \quad (8)$$

where K is also called activity product. In Eq. (8), the activity of water can be calculated from the osmotic coefficient by

$$\ln a_{H_2O} = -\phi \left(\frac{M_W}{1000} \right) \sum m_i \quad (9)$$

where ϕ is the osmotic coefficient. Assuming the electrolytes are completely ionized, the solubility of salt can be calculated by Eq. (8) if the activity coefficients of the ions in the electrolyte solution can be conveniently obtained. In this work, the modified MSA [12] is used to calculate the activity coefficients of electrolyte in saturated solution.

2.2. The modified mean spherical approximation

The primitive model of the MSA is an approximation method to solve Ornstein–Zernike integral equation. In the modified MSA, the expression for the activity coefficient is a sum of an electrostatic and a hard sphere term

$$\ln \gamma_i = \ln \gamma_i^{\text{elec}} + \ln \gamma_i^{\text{hs}} \quad (10)$$

The first term in Eq. (10) can be expressed as

$$\ln \gamma_i^{\text{elec}} = \frac{z_i e^2 L_i}{DkT} - \frac{P_n \sigma_i}{4\Delta} \left(\Gamma a_i + \frac{\pi}{12\Delta} \alpha^2 P_n \sigma_i^2 \right) \quad (11)$$

where

$$\alpha^2 = \frac{4\pi e^2}{DkT}$$

$$P_n = \frac{1}{\Omega} \sum_{k=1}^N \frac{\rho_k \sigma_k z_k}{1 + \Gamma \sigma_k}$$

$$\Omega = 1 + \left(\frac{\pi}{2\Delta} \right) \sum_{k=1}^N \frac{\rho_k \sigma_k^3}{1 + \Gamma \sigma_k}$$

$$\Delta = 1 - \xi_3$$

$$\xi_n = \frac{\pi}{6} \sum_{i=1}^N \rho_k \sigma_k^n, \quad n = 0, 1, 2, 3$$

$$a_i = \alpha^2 \frac{z_i - (\pi/2\Delta)\sigma_i^2 P_n}{2\Gamma(1 + \alpha\sigma_i)}$$

$$L_i = \frac{2\Gamma a_i / \alpha^2 - z_i}{\sigma_i}$$

The shielding parameter Γ can be obtained from

$$4\Gamma^2 = \alpha^2 \sum_{k=1}^N \rho_k \left(\frac{z_k - (\pi/2\Delta)\sigma_k^2 P_n}{1 + \Gamma\sigma_k} \right)^2 \quad (12)$$

Eq. (12) can be solved by a simple iterative procedure. $\Gamma = \kappa/2$ can be taken as the initial value, where

$$\kappa = \alpha \left(\sum_{i=1}^N \rho_k z_k^2 \right)^{1/2}$$

The expression for hard sphere term is obtained from equation of state for a mixture of hard spheres [14,15]

$$\ln \gamma_i^{\text{hs}} = -\ln \Delta + \frac{\pi P^{\text{hs}} \sigma_i^3}{6 kT} + E + 3F^2 G - F^3 H \quad (13)$$

where

$$E = \frac{3\xi_2 \sigma_i + 3\xi_1 \sigma_i^2}{\Delta} + \frac{9}{2} \frac{\xi_2^2 \sigma_i^2}{\Delta^2}$$

$$F = \frac{\xi_2 \sigma_i}{\xi_3}$$

$$G = \ln \Delta + \frac{\xi_3}{\Delta} - \frac{\xi_3^2}{2\Delta^2}$$

$$H = 2 \ln \Delta + \frac{\xi_3(2 - \xi_3)}{\Delta}$$

$$P^{\text{hs}} = \frac{6kT}{\pi} \left[\frac{\xi_0}{\Delta} + \frac{3\xi_1 \xi_2}{\Delta^2} + \frac{\xi_2^3(3 - \xi_3)}{\Delta^3} \right]$$

In the calculation, the anion diameters in Eq. (11)–(13) are constant, and the cation diameters are effective diameters of hydrated ions. The cation diameters can be calculated by

$$\sigma_+ = \sigma_0 - \lambda_1 \frac{I^{1/2}}{1 + I^{1/2}} - \lambda_2 I^2 \quad (14)$$

where σ_0 , λ_1 and λ_2 are the effective diameter parameters of the cations, and λ_2 is only present at high concentration. Their values can be obtained from Lu et al. [12].

If only one kind of anion exists in the mixed electrolyte solution, the effective diameter of a cation can be calculated from the following equation:

$$\sigma_M = \sigma_0 - \lambda_1 \left(\frac{I_{MA}^{1/2}}{1 + I_{MA}^{1/2}} \right) - \lambda_2 I_{MA}^2 \quad (15)$$

where σ_0 , λ_1 and λ_2 are the same as the effective diameter parameters of the cations and I_{MA} is the total ionic strength of electrolyte MA in the mixed electrolyte solution:

$$I_{MA} = \frac{1}{2}(m_M z_M^2 + m_A z_A^2) \quad (16)$$

where m_M and m_A are the total molalities of ions M and A in the solution, respectively.

If there are different anions in the solution, the effective diameter of cation M can be obtained from a linear mixing rule

$$\sigma_{M(\text{mix})} = \sum_{j=1} \sigma_{M(A_j)} X_{A_j} \quad (17)$$

where

$$X_{A_j} = \frac{m_A}{\sum_{j=1} m_{A_j}}$$

It should be pointed that the activity coefficient calculated from Eqs. (10)–(17) is based on molarity scale and in McMillan–Mayer (MM) reference state. Although better corrections [16,17] have been suggested, here we use equation proposed by Pailthorpe et al. [18] to change the activity coefficient into that in Lewis–Randall (LR) reference state.

$$\ln \gamma_{\pm}^{\text{LR}} = \ln \gamma_{\pm}^{\text{MM}} - \frac{\Pi \bar{V}_{\pm}}{RT} \quad (18)$$

where the osmotic pressure is calculated by $\Pi = \nu RT m M_W \phi / 1000 \bar{V}_W$. Here, \bar{V}_{\pm} and \bar{V}_W are the mean partial molar volume of the electrolyte and the partial molar volume of water, respectively. The partial molar volumes of electrolyte and water can be obtained from the density of electrolyte solution. In this work, the equation proposed by Novotuy and Sohnel [19] was adopted to express the densities of a single electrolyte solution.

Because literature data for the density of mixed electrolyte solutions are scarce, we use an ionic strength average from the densities of the corresponding single electrolyte solution

$$d_{\text{mix}}(I, T) = \sum_{i=1} d_i(I, T) y_i \quad (19)$$

where $d_i(I, T)$ is the density of single electrolyte i at ionic strength I and temperature T , and y_i the ion strength fraction. When the activity coefficient was obtained, the osmotic coefficient can be calculated using Gibbs–Duhem equation for ternary electrolyte solution. For simplicity, an approach proposed in previous work [12,13] can be employed to estimate the osmotic coefficients in mixed electrolyte solutions according to the ionic strength fraction y_j

$$\phi_{\text{mix}}(I, T) = \sum_j \phi_j(I, T) y_j \quad (20)$$

where $\phi_j(I, T)$ is the osmotic coefficient of the corresponding single electrolyte solution at temperature T and ionic strength I . It can be calculated using Gibbs–Duhem equation for binary solution.

3. Results and discussion

In this work, we assume that electrolytes in the solutions are completely ionized, and dielectric constants of electrolyte solutions are replaced with that of pure water. Such simplification is appropriate for the calculation of the activity coefficient of strong electrolyte. But if non-ionized electrolyte is neglected in the calculation of solid–liquid equilibria, the predicted value of solubility of salt is under-estimated. However, a part of the deviation in the calculation for the salt solubility will be canceled if the activity product is obtained from the salt solubility data in pure water. Of course, such an assumption is not suitable for weak electrolytes. In the calculation, the dielectric constant for water is obtained from [20]

$$D_{\text{H}_2\text{O}} = 87.74 - (4.0008 \times 10^{-1}t) + (9.398 \times 10^{-4}t^2) - (1.41 \times 10^{-6}t^3) \quad (21)$$

where $t = T - 298.15$ K.

In solution mixtures saturated with respect to salt $M_{\nu_M}A_{\nu_A} \cdot \nu_0\text{H}_2\text{O}$, the activity product of the salt must be equal to that in a pure saturated solution at the same temperature and pressure

$$K = (m_M^* \gamma_M^*)^{\nu_M} (m_A^* \gamma_A^*)^{\nu_A} (a_{\text{H}_2\text{O}}^*)^{\nu_0} \quad (22)$$

where superscript * denotes a pure salt solution property. Values of m_M^* and m_A^* for each salt are from the pure saturated solution, and the corresponding γ_M^* , γ_A^* and $a_{\text{H}_2\text{O}}^*$ were calculated using the modified MSA with parameters listed in Table 1 to maintain self-consistency.

The calculation for solubility of salt in mixed electrolyte aqueous solution starts with a given initial value of the solubility $m^{(0)}$. The activity coefficients of ions and the activity of water are then updated using Eqs. (9)–(12). A new solubility $m^{(1)}$ of salt is obtained from Eq. (8). If $|m^{(1)} - m^{(0)}| \leq 10^{-4}$, $m^{(1)}$ is the actual solubility of salt; if $|m^{(1)} - m^{(0)}| > 10^{-4}$, we adjust $m^{(0)}$ and repeat the calculation until $|m^{(n+1)} - m^{(n)}| \leq 10^{-4}$.

Table 1

Diameters of anions and effective diameter parameters of cations for electrolytes involved in this work [12]

Electrolyte	σ_- (nm)	Effective diameter parameters of cation			m_{max}
		σ_0 ($\times 10$ nm)	λ_1 ($\times 10$ nm)	λ_2 ($\times 10^4$ nm mol kg $^{-2}$)	
HCl	0.362	5.555	1.928	2.788	9.0
KCl	0.362	3.657	1.907	–	5.0
KNO ₃	0.230	3.292	4.226	–4.742	3.5
LiCl	0.362	5.281	1.789	1.547	19.2
Li ₂ SO ₄	0.285	6.738	4.254	–1.458	3.2
MgCl ₂	0.362	7.552	2.770	1.124	5.9
NaCl	0.362	4.371	2.266	–4.189	6.1
NaClO ₃	0.323	4.601	3.066	–2.497	3.0
NaNO ₃	0.230	4.732	3.030	–1.929	10.8
Na ₂ SO ₄	0.285	5.780	4.584	–0.795	2.0

Table 2

Predicted results of solid–liquid equilibria for mixed electrolyte aqueous solution at 298.15 K

Systems	Solid phase	NP	$\ln K$	I_m	ARD (%)	Reference
HCl + KCl	KCl	8	2.080	6.26	7.72	[21]
LiCl + Li ₂ SO ₄	Li ₂ SO ₄ ·H ₂ O	4	1.200	16.30	9.95	[22]
NaCl + MgCl ₂	NaCl	6	3.639	15.00	10.15	[23]
LiCl + KCl	LiCl	3	14.046	20.80	2.67	[24]
KCl + NaCl	KCl	4	2.080	7.28	10.56	[25]
KCl + NaCl	NaCl	4	3.639	7.28	3.41	[25]
NaClO ₃ + NaCl	NaCl	7	3.639	8.44	0.81	[26]
NaCl + NaNO ₃	NaCl	5	3.639	11.10	1.50	[25]
NaCl + NaNO ₃	NaNO ₃	4	2.502	10.90	3.47	[25]

The total predicted results of solid–liquid equilibria at 298.15 K for seven mixed electrolyte aqueous solutions are listed in Table 2. The sources of the experimental data for the solubilities are listed in the last column of Table 2. Also included in Table 2 are solid salts which are in equilibrium with the corresponding mixed electrolyte solutions, the maximum ion strengths (I_m) and the average relative deviations (ARDs) between the predicted and experimental values. The average relative deviation is defined as

$$\text{ARD} (\%) = \sum_{i=1}^{\text{NP}} \frac{|m^{\text{cal}} - m^{\text{exp}}|}{m^{\text{exp}}} \times 100 \quad (23)$$

From Table 2, one can see that the prediction accuracy is good for the seven mixed electrolyte systems. The total average ARD is 5.58% and the maximum ARD is 10.56% for aqueous KCl + NaCl system.

In Fig. 1, we compared the predicted and measured solubilities of NaCl in aqueous MgCl₂ solution at 298.15 K. Fig. 2 shows comparisons between the predicted and experimental value for the solubility of NaCl in aqueous NaClO₃ solution at 298.15 K. As shown in Figs. 1 and 2, the predicted curves of the solubility are in good agreement with the experimental data.

In order to test the prediction ability of our model for solid–liquid equilibria of mixed electrolyte systems at various temperatures, the solid–liquid equilibria for four mixed electrolyte aqueous solutions

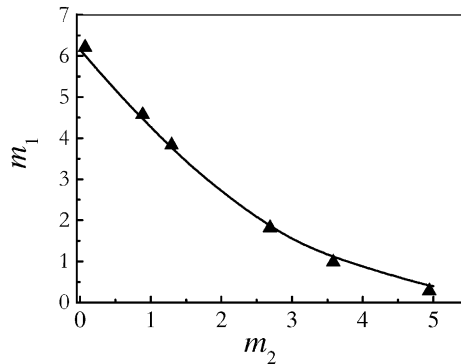


Fig. 1. Solubilities of NaCl (1) in aqueous MgCl₂ (2) solution at $T = 298.15$ K. Symbols stand for the experimental values and solid line refers to the predicted values.

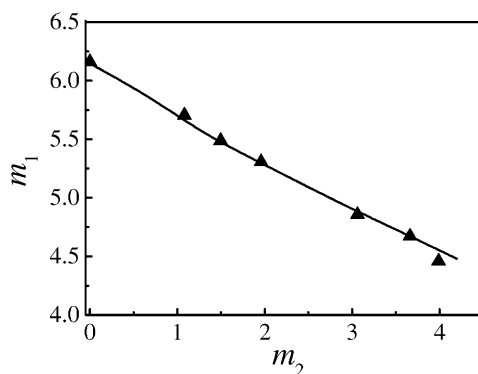


Fig. 2. Solubilities of NaCl (1) in aqueous NaClO₃ (2) solution at $T = 298.15$ K. Symbols stand for the experimental values and solid line refers to the predicted values.

Table 3

Predicted results of solid–liquid equilibria for mixed electrolyte aqueous solution at various temperatures

Systems	Solid phase	T (K)	NP	I_m	ARD (%)	Reference
NaCl + KCl	NaCl	303.15–353.15	12	8.77	2.62	[27]
KNO ₃ + NaCl	NaCl	293.15–364.15	17	24.40	4.36	[25]
NaCl + Na ₂ SO ₄	Na ₂ SO ₄	298.15–373.15	4	8.04	7.53	[28]
Li ₂ SO ₄ + Na ₂ SO ₄	Li ₂ SO ₄	273.15–318.15	12	12.2	6.42	[25]

at some temperatures other than 298.15 K are calculated only using the model parameters obtained at 298.15 K. Table 3 shows the predicted results of the solid–liquid equilibria for the four mixed electrolyte aqueous solutions at temperature range from 273.15 to 373.15 K. From Table 3, it can be seen that the average absolute deviation between the predicted and experimental values for the four mixed electrolyte systems is less than 8% although maximum ionic strength is up to 24.4 mol kg⁻¹. Comparisons between the predictions and experimental solubility data suggest that unlike semi-empirical models, the parameters in the modified MSA are insensitive to temperature. The deviation for symmetrical electrolyte mixture with common ions is better than those for asymmetrical electrolyte system. Our predicted results from the modified MSA are adequate for engineering application.

4. Conclusion

By taking the activity product of salt as a criterion of solid–liquid equilibria, the solubility of salts in seven mixed electrolyte aqueous solutions at 298.15 K are predicted. The activity coefficients of electrolyte and the activity of water in the solution are calculated from the modified mean spherical approximation method. The average absolute deviation between predicted results and experimental data published in literature is 5.58%. The solid–liquid equilibria for mixed electrolyte aqueous solutions are predicted at various temperatures using the parameters of single electrolyte obtained at 298.15 K. The predicted results show that the present method can predict the solid–liquid equilibria of mixed electrolyte aqueous

solution with good accuracy over a range of temperature. The calculation in this work is limited to ternary solutions, but applying the modified MSA to arbitrary multi-component electrolyte solutions is straightforward.

List of symbols

a	activity
d	density of solution (kg m^{-3})
D	dielectric constant
e	unit electronic charge (C)
I	ion strength (mol kg^{-1})
k	Boltzmann constant
K	equilibrium constant of dissociation; activity product of salt
m	molality (mol kg^{-1})
M	molecular weight (g mol^{-1})
NP	number of experimental point
P	pressure (Pa)
R	gas constant ($8.3144\text{J mol}^{-1} \text{K}^{-1}$)
T	absolute temperature (K)
V	partial molar volume ($\text{m}^3 \text{mol}^{-1}$) or volume of solution ($\text{m}^3 \text{mol}^{-1}$)
X	mole fraction of anion
y	ion strength fraction
z	valence of ion

Greek letters

γ	activity coefficient
Γ	shielding parameter
ν	stoichiometric coefficient
λ	parameter of effective diameter of cation (nm)
μ	chemical potential (J mol^{-1})
Π	osmotic pressure (Pa)
ρ	number density (nm^{-3})
σ	hard sphere diameter or effective diameter (nm)
ϕ	osmotic coefficient

Subscripts

A_j	anion
i, j, k	components i, j, k
L	liquid state
m	maximum
mix	mixed electrolyte solution
M	cation
S	solid state
W	water
\pm	ionic average

+	cation
–	anion

Superscripts

cal	calculated value
elec	electrostatic
exp	experimental value
hs	hard sphere
LR	Lewis–Randall reference state
MM	McMillan–Mayer reference state
0	standard state
*	pure salt solution

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