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Isopiestic Study of the Na₂CrO₄-H₂O System at 353.15 K: Prediction of the Solubility of Na₂CrO₄ in Aqueous NaOH Solutions

Wei Jin, †,‡ Shili Zheng,*,† Hao Du,† Hongbin Xu,† and Yi Zhang†

National Engineering Laboratory for Hydrometallurgical Cleaner Production Technology, Key Laboratory of Green Process and Engineering, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, People's Republic of China, and Graduate School of Chinese Academy of Sciences, Beijing 100049, People's Republic of China

The osmotic coefficients of the $Na_2CrO_4-H_2O$ system at 353.15 K were determined using isopiestic method ranging from 0.05 to 7.683 mol·kg⁻¹. The experimental osmotic coefficients were excellently correlated with Pitzer equation to obtain the interaction parameters, the activity coefficients, and the solubility product for Na_2CrO_4 . Furthermore, the solubility of Na_2CrO_4 in NaOH solutions at 353.15 K was predicted using binary parameters, and the predicted values agree with the experimental data, demonstrating the Pitzer equation is capable of optimizing the processes for the removal and recovery of sodium chromate in NaOH solutions at high temperature.

1. Introduction

Chromium-bearing sodium hydroxide solutions are of great importance in geochemistry, environmental treatment, and industrial processes.^{3,4} In alkaline solutions, Cr exists primarily as the highly soluble oxyanionic species, CrO_4^{2-} , which can lead to significant groundwater contamination as a result of release from rock or from human activity.⁵ Among all hazardous wastes, Cr is the third most frequently cited substance and among metallic substances is second only to Pb in cited frequency.⁶ Consequently, the aqueous chemistry of alkaline sodium chromate solutions is critical for optimizing numerous industrial treatments, 3,4 such as the basic dissolution of chromite ore, electroplating, wood pulp production, leather-tanning, and petroleum refining. Consequently, there is a significant requirement for a reliable, comprehensive, internally consistent set of thermodynamic properties for predicting the phase behavior of Cr in NaOH solutions.

Considerable research has been carried out on the solubility diagrams of NaOH-Na₂CrO₄-H₂O systems at various temperatures and concentrations. ^{7,8} To systematically correlate and predict the solid-liquid equilibrium, it has been necessary to determine the osmotic and activity coefficients. Due to the great flexibility and wide applicability, the isopiestic method is available for aqueous mixed salt solutions. On the basis of the isopiestic method, abundant experimental osmotic and activity coefficient data for the NaOH-H2O binary system with significant temperature and concentration ranges have been obtained, 9,10 while data for the Na₂CrO₄-H₂O system are only available at 298.15 K.11 However, the unit operations during chromium-bearing industrial processes mainly focus on the temperature ranging from 313.15 to 353.15 K,12 addressing the necessity for the determination of osmotic and activity coefficients for the Na₂CrO₄-H₂O system in such regions for theoretical and practical reasons. In this regard, the aim of this study is to measure the osmotic coefficients for the Na₂CrO₄-H₂O system from m = 0.05 to 7.683 mol·kg⁻¹ at 353.15 K using our specially designed isopiestic apparatus. The experimental data were then correlated with the Pitzer equation 13 to give the reliable parameters. The obtained parameters were employed and coupled with the parameters of the NaOH-H $_2$ O system previously reported to predict the solubility of Na $_2$ CrO $_4$ in NaOH aqueous solutions at 353.15 K.

2. Experimental Section

In this study, the isopiestic method is used to determine the osmotic coefficients of Na₂CrO₄ solutions. It is based on the principle that¹⁴ different solutions, when connected through the vapor space, approach equilibrium by transferring solvent due to the chemical potential gradients of the solvent in each solution. At equilibrium, the chemical potentials of the solvent in each solution are identical, which implies the equality of the solvent activity.

Experiments were conducted in a slightly modified version of the apparatus described by Zhou¹⁵ due to its favorable reliability and a fairly fast rate of equilibration. The apparatus mainly consisted of four small nickel cups for the reference standard solution samples and one big nickel cup for the sample being measured. The stock solution of Na₂CrO₄ and corresponding CaCl₂ reference standard solutions were prepared by dissolving Na₂CrO₄ and CaCl₂ reagents (both p.a. grade, Sigma-Aldrich) in high-purity Milli-Q water. The weight of the sample was precisely weighed, the mass of the sodium chromate solution was approximately 85 g, and the corresponding reference solutions were 1.0-1.5 g for each. Prior to each measurement, air was removed from the apparatus with a vacuum pump, and to prevent spattering of the solution during evacuation, the apparatus was connected to an empty beaker flask, which acted as an intermediate reservoir. Then the apparatus was put on a rotating platform in a thermostatic water bath at 353.15 K with a temperature precision of 0.1 K (SC-30A, Ninbo Xinzhi), and the rotating speed was set to 40 rpm to accelerate the equilibration.

The equilibrium was achieved when the differences between the mass fractions of reference solutions were less than 1%, and then the average of the molalities of the references was reported. Besides, the initial and final molalities of the samples and reference solutions were further analyzed by inductively

^{*} To whom correspondence should be addressed. E-mail: slzheng@ home.ipe.ac.cn. Fax: +86-10-62520910.

[†] Institute of Process Engineering, Chinese Academy of Sciences.

^{*} Graduate School of Chinese Academy of Sciences.

Table 1. Experimental Osmotic Coefficients of $NaOH-H_2O$ and $Na_2CrO_4-H_2O$ Systems and the Calculated Osmotic and Mean Ionic Activity Coefficients at 353.15 K

experiments	m (mol/kg)	CaCl ₂ (mol/kg)	Φ_{expt}	γ_{\pm}	Φ_{calc}	$\delta \times 100^a$
NaOH	0.50	0.3545	0.9030		0.9010	0.22
	1.00	0.6750	0.9222		0.9205	0.18
	3.00	1.7501	1.0560		1.0601	0.38
	4.50	2.4802	1.1834		1.1828	0.05
Na ₂ CrO ₄	0.05	0.0483	0.8147	0.5007	0.8112	0.44
	0.1	0.0930	0.7724	0.4187	0.7755	0.41
	0.2	0.1783	0.7392	0.3412	0.7389	0.04
	0.5	0.4030	0.6906	0.2502	0.6906	0.00
	0.8	0.6032	0.6749	0.2119	0.6740	0.14
	1	0.7282	0.6721	0.1966	0.6723	0.04
	1.2	0.8521	0.6766	0.1860	0.6762	0.07
	1.5	1.0312	0.6868	0.1756	0.6897	0.42
	2	1.3423	0.7287	0.1676	0.7277	0.14
	2.5	1.6584	0.7822	0.1671	0.7798	0.31
	3	1.9810	0.8435	0.1718	0.8426	0.11
	3.5	2.3152	0.9131	0.1811	0.9140	0.10
	4	2.6652	0.9918	0.1947	0.9930	0.12
	4.5	3.0303	1.0777	0.2130	1.0788	0.20
	5	3.4153	1.1724	0.2366	1.1709	0.13
	5.5	3.8023	1.2654	0.2665	1.2689	0.28
	6	4.2292	1.3763	0.3039	1.3727	0.26
	6.5	4.6541	1.4822	0.3508	1.4820	0.01
	7	5.1038	1.5965	0.4095	1.5967	0.01
	7.5	5.5720	1.7152	0.4831	1.7167	0.09
	7.683	5.7551	1.7630	0.5145	1.7620	0.06

^a $\delta \times 100 = ((\phi_{\text{calc}} - \phi_{\text{expt}})/(\phi_{\text{expt}})) \times 100.$

Table 2. Pitzer Parameters of Aqueous Binary Systems at 353.15 K

solute	$eta^{(0)}$	$eta^{(1)}$	C^{φ}
CaCl ₂ ^a	0.3204	1.7246	-0.0080 -0.0004 0.0052
NaOH ^b	0.0984	0.1610	
Na ₂ CrO ₄ ^c	0.0784	1.2282	

 a The parameters of CaCl $_2$ were correlated with the experimental data of Anathaswamy. 16 b The parameters of NaOH were taken from Christov. 18 c The parameter of Na $_2$ CrO $_4$ were regressed by this work.

coupled atomic plasma emission spectrometry (ICP-AES, 2400 type, Perkin-Elmer).

3. Results and Discussions

3.1. Experimental Data. Results of the isopiestic molalities of the equilibration experiments at 353.15 K are listed in Table 1. The experimental molality-based osmotic coefficient Φ of test solution was obtained using the fundamental equation for isopiestic equilibrium:

$$\Phi = \Phi^* \nu^* m^* / \nu m \tag{1}$$

where Φ^* is the osmotic coefficient of the reference solution in isopiestic equilibrium with the test solution, ν^* is the stoichiometric coefficient of the reference solution, m^* is the molality of the reference solution, ν is the stoichiometric coefficient of the test solution, and m is the molality of the test solution. The reference osmotic coefficient of CaCl₂ ($m_{\rm max}=6$ m) was calculated with the Pitzer model, in which the Pitzer parameters as shown in Table 2 are regressed from the experimental osmotic coefficient values of Anathaswamy. ¹⁶

To evaluate the reliability of our isopiestic apparatus, the experimental osmotic coefficients of NaOH-H₂O at 353.15 K are also reported in Table 1. We compared the experimental data with those calculated with the following Pitzer equation:

$$\Phi_{\text{NaOH}} = 1 - \frac{A_{\varphi} I^{1/2}}{1 + b I^{1/2}} + m(\beta_{\text{NaOH}}^{(0)} + \beta_{\text{NaOH}}^{(1)} e^{-\alpha I^{1/2}}) + m^2 C_{\text{NaOH}}^{\phi} (2)$$

in which the ionic strength I is given as

$$I = \frac{1}{2} \sum m_i z_i^2 \tag{3}$$

the Debye–Hückel parameter A_{ϕ} was taken as 0.4384 at 353.15 K calculated with Moller's equation, ^{17}b and α are the universal parameters generally considered to be 1.2 (kg mol $^{-1}$) $^{1/2}$ and 2.0 (kg mol $^{-1}$) $^{1/2}$, respectively, the temperature-dependent Pitzer parameters $\beta_{\text{NaOH}}^{(0)}$, $\beta_{\text{NaOH}}^{(1)}$, and C_{NaOH}^{ϕ} were taken from Christov 18 as shown in Table 2, m_i is the molality of species i in a multicomponent solution, and z_i is the valence of species i. The relative average deviation between experimental and calculated osmotic coefficients is 0.21%, indicating the apparatus and method are significantly reliable. Consequently, the osmotic coefficients of the Na₂CrO₄–H₂O system at 353.15 K were obtained and summarized in Table 1.

3.2. Correlation of the Na₂CrO₄-H₂O System with Pitzer Model. The experimental osmotic coefficients of Na₂CrO₄-H₂O were correlated with the Pitzer equation:

$$\Phi_{\text{Na}_2\text{CrO}_4} = 1 - \frac{2A_{\phi}I^{1/2}}{1 + bI^{1/2}} + \frac{4}{3}m(\beta_{\text{Na}_2\text{CrO}_4}^{(0)} + \beta_{\text{Na}_2\text{CrO}_4}^{(1)}e^{-\alpha I^{1/2}}) + \frac{2^{5/2}}{3}m^2C_{\text{Na}_2\text{CrO}_4}^{\phi}$$
(4)

The obtained Pitzer parameters are presented in Table 2. The calculated osmotic coefficients and their corresponding relative deviation are listed in Table 1; the comparison between the experimental and calculated osmotic coefficients of Na₂CrO₄–H₂O system as a function of molality is illustrated in Figure 1. The relative average deviation is 0.16% between experimental and calculated values. It is obvious that the experimental data are fully consistent with the correlated data, indicating the Pitzer model is valid for such a system ranging from 0.05 to 7.683 m. Therefore, the mean ionic activity coefficients of Na₂CrO₄ in pure water as a function of molality were calculated using the following Pitzer equation and presented in Table 1 and Figure 2:

$$\begin{split} \ln \gamma_{\pm,\mathrm{Na_2CrO_4}} &= -2A_\phi \bigg(\frac{I^{1/2}}{1 + bI^{1/2}} + \frac{2}{b} \ln(1 + bI^{1/2}) \bigg) + \\ &\frac{4}{3} m \bigg\{ 2\beta_{\mathrm{Na_2CrO_4}}^{(0)} + 2\frac{\beta_{\mathrm{Na_2CrO_4}}^{(1)}}{\alpha^2 I} \bigg[1 - \left(1 + \alpha I^{1/2} - \frac{1}{2}\alpha^2 I \right) \times \\ &\exp(-\alpha I^{1/2}) \bigg] \bigg\} + 2^{3/2} m^2 C_{\mathrm{Na_2CrO_4}}^{\phi} \ (5) \end{split}$$

As can be seen in Figure 1 and Figure 2, as the concentration increases, the osmotic and mean ionic activity coefficients initially significantly decrease and then increase monotonically. This phenomenon is consistent with other binary systems observed, and it is due to the electrolyte-dependent complex interaction among different ions and solvent. According to the classical aqueous electrolyte solution theory, ¹⁹ in dilute region, as the concentration increases, the ion association is facilitated; therefore the osmotic and activity coefficients are reduced. In the concentrated region, the γ and Φ are mainly determined by the hard sphere repulsive force; as the concentration increases,

Figure 1. Comparison between experimental and calculated osmotic coefficient values using the Pitzer equation for $Na_2CrO_4-H_2O$ at 353.15 K

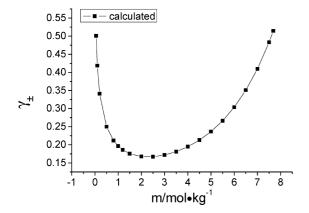


Figure 2. Calculated mean ionic activity coefficients for $Na_2CrO_4-H_2O$ system using the Pitzer equation at 353.15 K.

the aggravating repulsive force among ions inhibits the ion association, and therefore the γ and Φ increase.

3.3. Prediction of Na₂CrO₄ Solubility in Aqueous NaOH Solutions at 353.15 K. A typical application of activity coefficient models is to predict the solubility of a salt in an aqueous solution containing other salts. There are substantial phase diagrams for the Na₂CrO₄−H₂O²⁰ and NaOH−Na₂-CrO₄−H₂O^{7,8,21} systems ranging from 298.15 to 383.15 K. At 353.15 K, the solubility of Na₂CrO₄ is 7.683 mol⋅kg⁻¹ in the Na₂CrO₄−H₂O system, and the equilibrium solid phase is Na₂CrO₄ in both the Na₂CrO₄−H₂O and NaOH−Na₂CrO₄−H₂O systems. Therefore, the solid−liquid equilibrium is described as follows:

$$Na_2CrO_4 \xrightarrow{K_{sp}} 2Na^+ + CrO_4^{2-}$$
 (6)

The solubility product K_{sp,Na₂}CrO₄ is expressed as

$$K_{\text{sp,Na}_2\text{CrO}_4} = (a_{\text{Na}^+})^2 a_{\text{CrO}_4^{2^-}} = (m_{\text{Na}^+})^2 m_{\text{CrO}_4^{2^-}} (\gamma_{\pm,\text{Na}_2\text{CrO}_4})^3$$
(7)

In our ternary system, the $m_{\rm Na^+}$ is given by

$$m_{\text{Na}^+} = m_{\text{NaOH}} + 2m_{\text{Na}_2\text{CrO}_4}$$
 (8)

From our experimental activity coefficient data for Na_2CrO_4 aqueous binary system, the $K_{sp,Na_2}CrO_4$ is 247.06 at 353.15 K calculated using eq 7. Consequently, by employing the binary Pitzer parameters in Table 2, the solubility of Na_2CrO_4 in aqueous NaOH solutions at 353.15 K was predicted using the

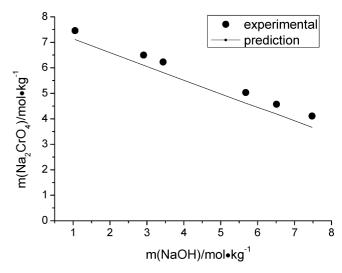


Figure 3. Comparison between experimental and predicted solubility of Na_2CrO_4 in $NaOH-H_2O$ at 353.15 K. (Experimental data were taken from Tou^{-7})

Pitzer model, in which $\gamma_{\pm, Na_2}CrO_4$ was obtained using the following equation:

$$\begin{split} \ln \gamma_{\pm,\mathrm{Na_2CrO_4}} &= -2A_\phi \bigg(\frac{I^{1/2}}{1 + bI^{1/2}} + \frac{2}{b} \ln(1 + bI^{1/2}) \bigg) + \\ & \frac{4}{3} m_{\mathrm{NaOH}} [B_{\mathrm{NaOH}} + (m_{\mathrm{NaOH}} + 2m_{\mathrm{Na_2CrO_4}}) C_{\mathrm{NaOH}}] + \\ & \bigg(\frac{2}{3} m_{\mathrm{NaOH}} + \frac{8}{3} m_{\mathrm{Na_2CrO_4}} \bigg) [B_{\mathrm{Na_2CrO_4}} + (m_{\mathrm{NaOH}} + 2m_{\mathrm{Na_2CrO_4}}) \times \\ & C_{\mathrm{Na_2CrO_4}}] + m_{\mathrm{NaOH}} (m_{\mathrm{NaOH}} + 2m_{\mathrm{Na_2CrO_4}}) \times \\ & \bigg(2B'_{\mathrm{NaOH}} + \frac{4}{3} C_{\mathrm{NaOH}} \bigg) + m_{\mathrm{Na_2CrO_4}} \times \\ & (m_{\mathrm{NaOH}} + 2m_{\mathrm{Na_2CrO_4}}) \bigg(2B'_{\mathrm{Na_2CrO_4}} + \frac{4}{3} C_{\mathrm{Na_2CrO_4}} \bigg) \end{split}$$
 (9)

in which

$$B_{\rm MX}(I) = \beta_{\rm MX}^{(0)} + \frac{2\beta_{\rm MX}^{(1)}}{\alpha^2 I} [1 - (1 + \alpha I^{1/2}) e^{-\alpha I^{1/2}}]$$
 (10)

$$C_{\rm MX} = \frac{C_{\rm MX}^{\phi}}{2|z_{\rm MZY}|^{1/2}} \tag{11}$$

$$B' = \frac{\partial B}{\partial I} \tag{12}$$

 B_{MX} and C_{MX} are the second and third virial coefficients in the excess Gibbs free energy, respectively, z_{M} is the cation valence, and z_{X} is the anion valence.

The comparison between experimental and predicted solubility is plotted in Figure 3. The deviations are relatively high, and increased with an increase in the concentration of the multicomponent solution. This result may be due to the relatively large contribution of mixed ionic interaction in the NaOH—Na₂CrO₄—H₂O system. Because the Pitzer model is applicable below 6 mol·kg⁻¹, it is difficult for the Pitzer model to predict the solubility at higher concentration. The concentration level of our ternary system was much higher than 6 mol·kg⁻¹. However, as can be seen in Figure 3, the predicted values are consistent with the average relative deviation of 7.52% in the experimental values below a NaOH concentration of 8.0 mol·kg⁻¹, indicating a satisfactory prediction has been achieved. As a whole, the application of the Pitzer equation to

predict the solubility of Na₂CrO₄ in aqueous NaOH solutions at 353.15 K using binary parameters is considered to be successful.

4. Conclusions

The osmotic coefficients of the Na₂CrO₄—H₂O system at 353.15 K ranging from 0.05 to 7.683 mol·kg⁻¹ were determined with isopiestic method. The reliability of the data obtained was validated by comparison with experimental results for NaOH solutions from studies previously reported.

The experimental osmotic coefficients were excellently correlated with the Pitzer equation. Thus, the interaction parameters, the mean ionic activity coefficients, and $K_{\rm sp,Na_2}CrO_4$ were obtained. Furthermore, the solubility of Na₂CrO₄ in NaOH solutions at 353.15 K was predicted using binary parameters. The predicted values are consistent with the average relative deviation of 7.52% of the experimental values, demonstrating the Pitzer equation is capable of optimizing the processes for the removal and recovery of sodium chromate in NaOH solutions at high temperature.

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Nomenclature

a = activity

 $A_{\Phi} = \text{Debye-H\"{u}ckel parameter}$

b = closet approach parameter in the Debye-Hückel term

 $B_{\rm MX} = {\rm second\ virial\ coefficient\ in\ the\ excess\ Gibbs\ free\ energy}$

 $C_{\rm MX}$ = third virial coefficient in the excess Gibbs free energy

 $C_{\rm MX}^{\phi}$ = Pitzer ion-interaction parameter

I = ionic strength

 $K_{\rm sp} =$ solubility product of solute in aqueous solutions

m = molality of test solution at equilibrium (mol/kg H₂O)

 $m_i = \text{molality of species } i \text{ (mol/kg H}_2\text{O)}$

 m^* = molality of reference solution at equilibrium (mol/kg H₂O)

 z_i = valence of species i

 $z_{\rm M} = {\rm cation \ valence}$

 $z_{\rm X}$ = anion valence

Greek Letters

 α = nonlinear parameter for second virial coefficient in the Pitzer equation

 $\beta_{\rm MX}^{(0)}, \beta_{\rm MX}^{(1)} = \text{Pitzer ion-interaction parameters}$

 γ_{\pm} = mean ionic activity coefficient

 ν , ν^* = stoichiometric coefficient for the samples and reference solution, respectively

 ϕ , ϕ * = osmotic coefficient for the test and reference solution, respectively

Subscripts

M = cation

X = anion

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