

DETERMINATION OF PITZER PARAMETERS FOR SODIUM BENZOATE AT 298.15 K

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Abstract

Sodium benzoate is a salt widely used in industry as preserver. Its activity coefficients in aqueous solutions can be estimated by Pitzer theory, once the necessary parameters are known. Electromotive force (emf) measurements on galvanic cells without liquid junction have been used for the determination of mean activity coefficients of sodium chloride in the ternary system sodium chloride + sodium benzoate (NaB) + water. Pitzer theory has been applied to this system and those data enabled the evaluation of the specific parameters for sodium benzoate at 298.15 K by linear regression. The values obtained for $\beta_{\text{NaB}}^{(0)}$ and $\beta_{\text{NaB}}^{(1)}$ parameters, respectively 0.177 kg mol⁻¹ and 0.34 kg mol⁻¹ are close to those published already for other carboxylic acid salts.

Keywords: sodium benzoate; activity coefficients; Pitzer theory; potentiometry

Introduction

Sodium benzoate is widely used in industry, primarily as preserver in foods and paper, and its thermodynamic properties, including activity coefficients, are still not well known.

In 1923, P. Debye and H. Hückel [1] presented a theory on inter-ionic attraction effects. From this theory a equation for a mean activity coefficient for a MX electrolyte (γ_{MX}) could be obtained:

$$\log \gamma_{\text{MX}} = -\frac{A|z_{\text{M}}z_{\text{X}}|I^{1/2}}{1 + BaI^{1/2}} \quad (1)$$

where A and B are constants of the Debye-Hückel theory, a is an ion size parameter related to the hydrated ions and z means ionic charge for cation M and anion X; $A = 0.5108 \text{ mol}^{-1/2} \text{ kg}^{1/2}$ and $B = 0.3286 \text{ cm}^{-1} \text{ mol}^{-1/2} \text{ kg}^{1/2}$ at 298.15 K. This equation has been used in prediction of activity coefficients values in aqueous solutions with good results for $I < 0.1 \text{ M}$, for 1:1 salts.

After the Debye and Hückel work, other researchers looked at the problem of calculating activity coefficients of an electrolyte in mixed solutions. Harned [2] presented expressions where the coefficients are specific to a given mixture and are functions of the temperature and ionic strength, but not of the composition [3]. Guggenheim equations [4] are only applicable to 1:1 electrolytes. Scatchard equations [5, 6] were cumbersome and the interaction parameters had physical significance not clear [3].

Pitzer theory [7-10] has been applied to the activity coefficients calculation of many salts of different valences, in aqueous solutions with ionic strength up to near saturation. The theory is based on a semi-empirical equations system for describing the non-ideal behaviour of electrolyte solutions.

Pitzer equations include terms for both electrostatic and short-range forces. However, the distinction between these terms is a consequence of the selected expression by Pitzer for the electrostatic contribution [7]. Pitzer obtained a general expression for the excess free energy of the mixed electrolyte:

$$\frac{G^{ex}}{RT} = w_w f(I) + \frac{1}{w_w} \sum_i \sum_j \lambda_{ij}(I) n_i n_j + \frac{1}{w_w^2} \sum_i \sum_j \sum_k \mu_{ijk} n_i n_j n_k \quad (2)$$

where $f(I)$ is a function of the ionic strength, temperature and nature of the solvent, corresponding to the electrostatic term of Debye and Hückel, expressing the effect of the long range electrostatic forces; $\lambda_{ij}(I)$ is also a function of the ionic strength and it considers the short distance binary interactions between the species i and j ; the μ_{ijk} term, assumed independent on the ionic strength, represents the ionic triple interactions. From the derivation of the equation (2) in order to n_i and for only one solute MX in solution, its activity coefficient is given by:

$$\ln \gamma_{MX} = |z_M z_X| f^\gamma + m \left(\frac{2v_M v_X}{v} \right) B_{MX}^\gamma + m^2 \left[\frac{2(v_M v_X)^{3/2}}{v} \right] C_{MX}^\gamma \quad (3)$$

where m is the molality of the electrolyte MX, v_M and v_X are the number of ions M and X per molecule, with electric charges z_M and z_X , respectively. Other quantities have the form

$$v = v_M + v_X \quad (4)$$

$$f^\gamma = -A_\phi \left[\frac{I^{1/2}}{(1 + bI^{1/2})} + \frac{2}{b} \ln(1 + bI^{1/2}) \right] \quad (5)$$

$$B_{MX}^\gamma = 2\beta^{(0)} + \left(\frac{2\beta^{(1)}}{\alpha^2 I} \right) \left[1 - \exp(-\alpha I^{1/2}) (1 + \alpha I^{1/2} - \frac{1}{2} \alpha^2 I) \right] \quad (6)$$

$$C_{MX}^\gamma = \frac{3}{2} C_{MX}^\phi \quad (7)$$

A_ϕ is the Debye-Hückel coefficient for the osmotic coefficient ($0.3915 \text{ mol kg}^{-1}$, at 298.15 K in water, [10]); b and α are two adjustable parameters with the values $b=1.2$ and $\alpha=2.0$ for 1:1 studied electrolytes, according to [7]. The $\beta^{(0)}$ and $\beta^{(1)}$ parameters define the second virial coefficient representing specific interaction parameters for the pure electrolytes, whereas C^ϕ defines the third virial coefficient, which usually is very small and sometimes negligible. All these parameters, $\beta^{(0)}$, $\beta^{(1)}$ and C^ϕ have been tabulated for many electrolytes [10].

The mean activity coefficient of the solute MX, in a mixture of electrolytes, is given by [10]:

$$\begin{aligned} \ln \gamma_{MX} = & |z_M z_X| \left[F + \frac{v_M}{v} \sum_a m_a [2B_{Ma} + ZC_{Ma} + 2(v_X/v_M)\Phi_{Xa}] + \frac{v_X}{v} \sum_c m_c [2B_{cX} + \right. \\ & \left. + ZC_{cX} + 2(v_M/v_X)\Phi_{Mc}] + \sum_c \sum_a m_c m_a \frac{1}{v} [2v_M z_M C_{ca} + v_M \Psi_{Mca} + v_X \Psi_{caX}] + \right. \\ & \left. + \sum_{c < c'} \sum_{a < a'} m_c m_{c'} \frac{v_X}{v} \Psi_{cc'X} + \sum_{a < a'} \sum_{c < c'} m_a m_{a'} \frac{v_M}{v} \Psi_{Maa'} + 2 \sum_n m_n (v_M \lambda_{nM} + v_X \lambda_{nX}) / v \right] \quad (8) \end{aligned}$$

Indices c and c' apply to all cations while a and a' cover all anions, and

$$Z = \sum_c m_c z_c = \sum_a m_a |z_a| \quad (9)$$

$$F = f^\gamma + \sum_c \sum_a m_c m_a B_{ca}' + \sum_{c < c'} \sum_{a < a'} m_c m_{c'} \Phi_{cc'}' + \sum_{a < a'} \sum_{c < c'} m_a m_{a'} \Phi_{aa'}' \quad (10)$$

$$B_{MX} = \beta_{MX}^{(0)} + \beta_{MX}^{(1)} \frac{1}{2I} [1 - (1 + 2\sqrt{I}) \exp(-2\sqrt{I})] \quad (11)$$

$$B_{MX}' = \frac{\beta_{MX}^{(1)}}{2I^2} [-1 + (1 + 2\sqrt{I} + 2I) \exp(-2\sqrt{I})] \quad (12)$$

$$C_{MX} = \frac{C_{MX}^\phi}{2|z_M z_X|^{1/2}} \quad (13)$$

Parameters Φ , Φ' and Ψ arise from additional combinations of the individual second and third virial coefficients, and they have been extensively tabulated [10]. Parameters with apostrophe correspond to the derivative of the parameter in order to ionic strength. Namely Φ' represents additional terms for higher order electrostatic effects for interactions between single and double or higher charged ions. According to Yang and Pitzer [11] these terms are negligible when only single charged ions are involved in interactions, so these terms were omitted in this work. Parameters $\lambda_{n...}$ are related to the interactions between molecule, n , and an ion.

The equation for the calculation of the activity coefficient of a single cation, M, in a mixed electrolyte [10] is as follows:

$$\begin{aligned} \ln \gamma_M = & z_M^2 F + \sum_a m_a (2B_{Ma} + ZC_{Ma}) + \sum_c m_c (2\Phi_{Mc} + \sum_a m_a \Psi_{Mca}) + \sum_{a' < c'} \sum_{a'' < c''} m_a m_{a'} \Psi_{Maa'} + \\ & + z_M \sum_c \sum_a m_c m_a C_{ca} + 2 \sum_n m_n \lambda_{nM} + z_M \left[\sum_c m_c \frac{\lambda_{cc}}{z_c} - \sum_a \frac{m_a \lambda_{aa}}{|z_a|} + \right. \\ & \left. + \frac{3}{2} \sum_c \sum_a m_c m_a \left(\frac{\mu_{cca}}{z_c} - \frac{\mu_{caa}}{|z_a|} \right) \right] \end{aligned} \quad (14)$$

For an anion X, a corresponding expression is achieved by interchanging X for M, a for c, and c for a throughout:

$$\begin{aligned} \ln \gamma_X = & z_X^2 F + \sum_c m_c (2B_{Xc} + ZC_{Xc}) + \sum_a m_a (2\Phi_{Xa} + \sum_c m_c \Psi_{Xac}) + \sum_{c' < c''} \sum_{a' < a''} m_c m_{c'} \Psi_{Xcc'} + \\ & + |z_X| \sum_c \sum_a m_c m_a C_{ca} + 2 \sum_n m_n \lambda_{nX} + |z_X| \left[\sum_a m_a \frac{\lambda_{aa}}{|z_a|} - \sum_c \frac{m_c \lambda_{cc}}{z_c} + \right. \\ & \left. + \frac{3}{2} \sum_c \sum_a m_c m_a \left(\frac{\mu_{aac}}{|z_a|} - \frac{\mu_{acc}}{z_c} \right) \right] \end{aligned} \quad (15)$$

In equations (14) and (15) λ_{aa} (or λ_{cc}) parameters represent interactions between two ions of the same electrical charge signal, and μ parameters are related to triple interactions between two ions of the same electrical charge signal and one ion of different signal charge.

The application of the Pitzer theory demands the knowledge of specific parameters for the electrolytes [7]. However, sodium benzoate parameters are not available in the literature.

Experimental

Reagents

Silver nitrate p.a. (Merck) and sodium hydroxide p.a. (Merck) were used to prepare silver oxide for the preparation of the silver/silver chloride electrodes. Sodium benzoate (99% Aldrich) and potassium chloride (99+%, Aldrich) were used in the preparation of pure sodium chloride solutions and sodium chloride+sodium benzoate mixed solutions. All solutions were prepared with deionized water with specific conductivity of $1.2 \mu \text{ S cm}^{-1}$.

Apparatus

A thermostated water bath was used and the temperature controlled at $298.15 \pm 0.05 \text{ K}$ with a Pt 100 probe, calibrated by Instituto de Ciéncia Aplicada e Tecnologia (ICAT), Lisbon. The potential was measured with a potentiometer, Hewlett-Packard, model HP 3458A.

A glass Na^+ selective electrode (I.S.E.), ABB Kent-Taylor, and silver-silver chloride electrodes of the thermo-electrolytic type, prepared as described by Bates [12], were used in cells. The cells vessels were H type.

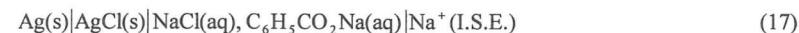
Procedure

For the solution preparation, sodium chloride was dried at 383.15 K for several hours, usually overnight, and cooled in a desiccator over blue gel before weighing. Sodium benzoate was dried at 343.15 K , to avoid decomposition, for as long as for sodium chloride. Solutions were prepared by weight and corrected to vacuum at 293.15 K .

In order to check the response of the electrodes, readings were taken for solutions of NaCl, with molalities ranging from 0.01 to 2 mol kg^{-1} . After the calculation of the activity coefficient, γ_{NaCl}^0 , using the Pitzer equation (3) with the parameters values for NaCl from Pitzer [10], a plot of the emf, E, against $\log(m_{\text{NaCl}}^0 \gamma_{\text{NaCl}}^0)$ showed a straight line with slope 118.45 mV , very close to the expected value, according to the Nernst equation.

Prior to each utilisation, silver/silver chloride electrodes were tested for their potential differences and were rejected if these were greater than 0.1 mV .

Series of solutions were made with the molality of Na^+ identical in both cells, (16) and (17), in order to reduce the asymmetry potential [13].



Potential readings were recorded when the variation was lower than 0.05 mV for at least 5 minutes, which would take, generally, around 20 minutes.

Results and discussion

Electromotive forces (emf) were measured in cells (16) and (17). The differences, ΔE , are shown in table 1 where γ_{NaCl}^0 is the activity coefficient of NaCl in cell (16), calculated by equation (3), and $\ln \gamma_{\text{NaCl}}(\text{exp})$ is the experimental value for the mean activity coefficient of NaCl in the mixed solution, given by [14]:

$$\ln \gamma_{\text{NaCl}}(\text{exp}) = \ln 10 \left[\frac{\Delta E}{2k} - \frac{1}{2} (\log m_{\text{Na}} + \log m_{\text{Cl}}) + \log m_{\text{NaCl}}^0 + \log \gamma_{\text{NaCl}}^0 \right] \quad (18)$$

where k is the Nernst factor, m refers to molality and the superscript 0 to a pure solution.

The value of $\ln \gamma_{\text{NaCl}}$, given by Pitzer model, equation (8), without the interaction parameters for neutral molecules for the mixed solutions is:

$$\begin{aligned} \ln \gamma_{\text{NaCl}}(\text{calc}) = & f^\gamma + m_{\text{Na}} m_{\text{Cl}} B'_{\text{NaCl}} + m_{\text{Na}} m_{\text{B}} B'_{\text{NaB}} + B_{\text{NaCl}} (m_{\text{Na}} + m_{\text{Cl}}) + \\ & + m_{\text{B}} B_{\text{NaB}} + 2 m_{\text{Na}} m_{\text{B}} C_{\text{NaB}} + m_{\text{B}} \theta_{\text{ClB}} + m_{\text{Na}} C_{\text{NaCl}} (m_{\text{Na}} + 2 m_{\text{Cl}}) + \\ & + \frac{1}{2} \Psi_{\text{Na,B,Cl}} (m_{\text{Na}} m_{\text{B}} + m_{\text{Cl}} m_{\text{B}}) \end{aligned} \quad (19)$$

with f^r , B and B' given by equations (5), (11) and (12), respectively. θ is the second virial coefficient for the mixture, C is the third virial coefficient for the pure electrolyte and Ψ for the mixed solution. Index B refers to benzoate anion.

Table 1 - Experimental natural logarithm activity coefficients for NaCl in pure and mixed solutions. ΔE corresponds to potential difference measured between cell (16) with aqueous pure NaCl solutions and cell (17) with mixed solutions (NaCl + sodium benzoate).

Pure NaCl solutions		Mixed solutions NaCl + sodium benzoate (NaB)		$\ln \gamma_{\text{NaCl}}$	$\Delta E / \text{mV}$
$m_{\text{NaCl}} / \text{mol kg}^{-1}$	γ_{NaCl}^0	$m_{\text{NaCl}} / \text{mol kg}^{-1}$	$m_{\text{NaB}} / \text{mol kg}^{-1}$		
0.0999	0.7769	0.0099	0.0899	-0.2366	-58.17
		0.0299	0.0699	-0.2492	-30.42
		0.0499	0.0499	-0.2526	-17.43
0.0997	0.7770	0.0099	0.0898	-0.2298	-58.16
		0.0199	0.0798	-0.2369	-40.58
		0.0398	0.0598	-0.2436	-23.70
		0.0599	0.0399	-0.2495	-12.92
0.1996	0.7323	0.0799	0.1199	-0.3069	-23.25
		0.0998	0.0998	-0.3059	-17.51
0.1997	0.7322	0.0200	0.1800	-0.2884	-57.75
		0.0599	0.1399	-0.2986	-30.10
		0.0988	0.0988	-0.3033	-17.77
0.2989	0.7075	0.0299	0.2697	-0.3150	-57.36
		0.0899	0.2099	-0.3207	-29.35
		0.1495	0.1495	-0.3290	-16.38
0.2999	0.7073	0.0299	0.2695	-0.2997	-56.75
		0.0599	0.2395	-0.3143	-39.65
		0.1197	0.1795	-0.3250	-22.43
		0.1499	0.1499	-0.3310	-16.91
		0.1789	0.1193	-0.3420	-13.07
0.3992	0.6911	0.0385	0.3461	-0.3396	-59.54
		0.0798	0.3194	-0.3327	-39.50
		0.1593	0.2389	-0.3384	-22.09
		0.1995	0.1995	-0.3457	-16.65
0.3995	0.6911	0.0399	0.3598	-0.3277	-56.99
		0.1198	0.2796	-0.3348	-29.13
		0.1999	0.1999	-0.3481	-16.64
0.4963	0.6799	0.0994	0.3978	-0.3566	-39.80
		0.1993	0.2989	-0.3458	-21.32
		0.2517	0.2517	-0.3707	-16.31

Table 1 - continuation

0.4997	0.6796	0.0499	0.4499	-0.3277	-56.13
		0.1499	0.3497	-0.3405	-28.54
		0.2495	0.2495	-0.3563	-16.29
0.5996	0.6713	0.0599	0.5397	-0.3352	-55.90
		0.1799	0.4197	-0.3496	-28.39
		0.2987	0.2987	-0.3689	-16.45
0.5997	0.6713	0.0598	0.5385	-0.3443	-56.52
		0.2396	0.3593	-0.3399	-20.61
0.6999	0.6652	0.0697	0.6274	-0.3353	-55.62
		0.2094	0.4887	-0.3485	-28.00
		0.3499	0.3499	-0.3670	-15.70
0.9972	0.6556	0.1998	0.7993	-0.3410	-37.10
		0.0999	0.8999	-0.3248	-53.69
0.9988	0.6555	0.2994	0.6985	-0.3541	-27.05
		0.4999	0.4999	-0.3794	-15.13
		0.1497	1.3473	-0.3017	-52.73
1.4966	0.6555	0.4489	1.0476	-0.3252	-25.73
		0.7475	0.7475	-0.3513	-14.00
		0.2935	1.1740	-0.3369	-35.05
1.4987	0.6556	0.5980	0.8971	-0.3115	-17.99

For each mixed solution, the difference between experimental $\ln \gamma_{\text{NaCl}}$ (exp), given by equation (18), and calculated $\ln \gamma_{\text{NaCl}}$ (calc), given by equation (19), with the unknown Pitzer parameters made zero, was calculated. A linear regression procedure with a spreadsheet (Microsoft Excel 2000) enabled to obtain parameters for sodium benzoate, shown in table 2.

Table 2 -Pitzer parameters for sodium benzoate obtained by linear regression analysis from the adjustment of calculated $\ln \gamma_{\text{NaCl}}$, equation (19) with the unknown Pitzer parameters made zero, to experimental $\ln \gamma_{\text{NaCl}}$ given by equation (18).

Parameters / kg mol^{-1}	Standard Error	Statistical Parameters
$\beta_{\text{NaB}}^{(0)}$	0.177	$R^2 = 0.982$
$\beta_{\text{NaB}}^{(1)}$	0.34	$\sigma = 0.00830$

R^2 - correlation coefficient; σ - standard error

Thus, from equations (18) and (19) the unknown parameters $\beta^{(0)}$ and $\beta^{(1)}$ for sodium benzoate could be determined, but this procedure does not allow the evaluation of θ and $\beta^{(0)}$ separately. However, the mixing parameter θ is expected to be very small and a value can be assigned to $\beta^{(0)}$ on the assumption that $\theta = 0$. The third virial coefficients, C_{NaB} and $\psi_{\text{Na,B,Cl}}$, were also included in the calculations and the variable estimates were smaller than their standard error, so their values were taken as zero. With the Pitzer coefficients for sodium benzoate thus obtained and shown in table 2, $\ln \gamma_{\text{NaCl}}(\text{calc})$ could be calculated and compared to $\ln \gamma_{\text{NaCl}}(\text{exp})$. Figure 1 shows these differences for the various solutions.

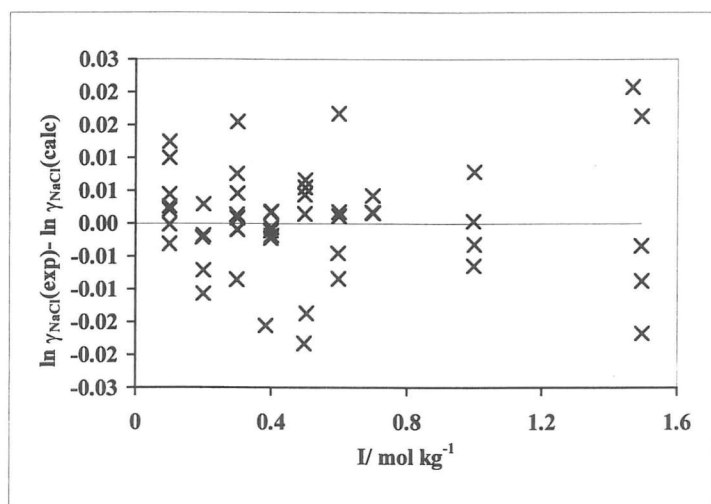


Figure 1. Differences between experimental and calculated $\ln \gamma_{\text{NaCl}}$ given respectively by equations (18) and (19), when Pitzer parameters for sodium benzoate are included in equation (19).

Figures 2 and 3 show the variation of $\ln \gamma_{\text{NaCl}}$ and $\ln \gamma_{\text{NaB}}$, respectively, with the ionic strength for mixed solutions of NaCl and NaB. Lines (a) and (b) represent the values for the limit situations, when only one electrolyte is present. $\ln \gamma_{\text{NaCl}}$ has been calculated by equation (19) and $\ln \gamma_{\text{NaB}}$ by an identical equation for sodium benzoate:

$$\begin{aligned} \ln \gamma_{\text{NaB}} = & f^{\gamma} + m_{\text{Na}} m_{\text{Cl}} B_{\text{NaCl}}^{\gamma} + m_{\text{Na}} m_{\text{B}} B_{\text{NaB}}^{\gamma} + B_{\text{NaB}} (m_{\text{Na}} + m_{\text{B}}) + \\ & + m_{\text{Cl}} B_{\text{NaCl}} + 2 m_{\text{Na}} m_{\text{Cl}} C_{\text{NaCl}} + m_{\text{Cl}} \theta_{\text{B,Cl}} + m_{\text{Na}} C_{\text{NaB}} (m_{\text{Na}} + 2 m_{\text{B}}) + \\ & + \frac{1}{2} \psi_{\text{Na,B,Cl}} (m_{\text{Na}} m_{\text{Cl}} + m_{\text{Cl}} m_{\text{B}}) \end{aligned} \quad (20)$$

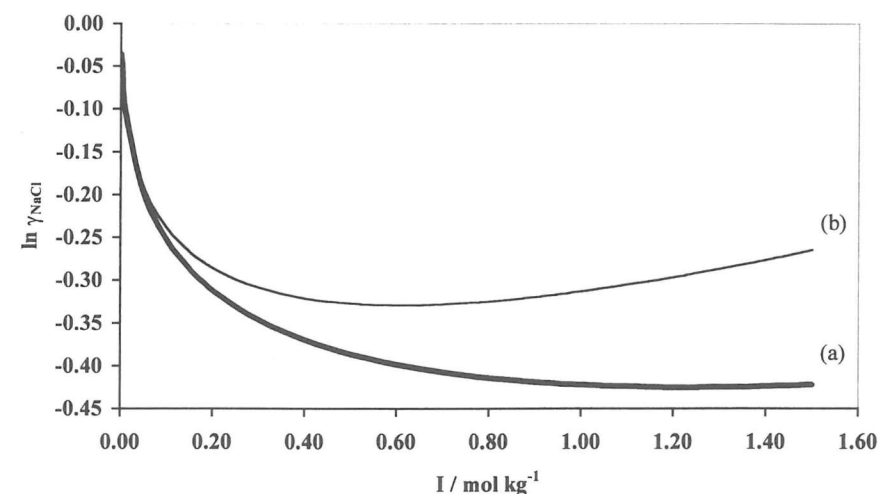


Figure 2. Variation of $\ln \gamma_{\text{NaCl}}$, given by equation (19), with ionic strength for the system NaCl + NaB + H₂O, when: (a) $m_{\text{NaB}}=0$; (b) $m_{\text{NaCl}}=0$.

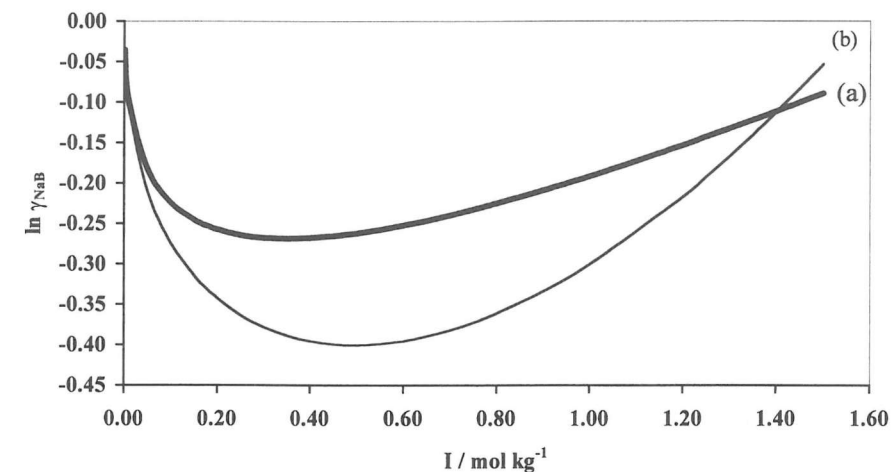


Figure 3. Variation of $\ln \gamma_{\text{NaB}}$ given by equation (20), with ionic strength for the system NaCl + NaB + H₂O, when: (a) $m_{\text{NaCl}}=0$; (b) $m_{\text{NaB}}=0$.

Conclusions

Theoretical Pitzer equation for $\ln \gamma_{\text{NaCl}}$ of sodium chloride in the electrolyte solution of cell (17) was used to fit the experimental data for $\ln \gamma_{\text{NaCl}}$ and the specific parameters for sodium benzoate were obtained by linear regression. This method has already been used in the determination of Pitzer parameters values of other salts [14]. The values thus obtained ($\beta_{\text{BNa}}^{(0)} = 0.177 \text{ kg mol}^{-1}$ and $\beta_{\text{BNa}}^{(1)} = 0.34 \text{ kg mol}^{-1}$) are close to those published already for other carboxylic acid salts [10].

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