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Equation for solvent activity correlation in ternary solutions with electrolyte and molecular components



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ARTICLE INFO

Article history: Received 26 August 2014 Received in revised form 15 December 2014 Accepted 28 December 2014 Available online 30 December 2014

Keywords: Solvent activity Ternary solutions Experimental data correlation

1. Introduction

Thermodynamics of solution is characterized by its ability to describe the mixing effects through the use of partial molar properties. For example, the chemical potential of a given component in solution is evaluated from the partial molar excess Gibbs free energy (RTln γ_i), which dependence on composition can be described by one of the many correlation equations available. The parameters of such equations are calculated starting from the correlation of the experimental dependence of the excess Gibbs free energy on composition.

A different situation arises when only one component, the solvent (1), is volatile, while the solutes (2, 3) can be considered non volatile (electrolytes, polysaccharides, amino acids, etc.). In these cases, the activity of the solvent can be evaluated directly from the corresponding solution vapor pressure and thus the experimental dependence of the solvent activity on composition $a_1(m_2,m_3)$ is obtained. To describe these systems, binary or ternary, it is usual to employ polynomial expressions [1–11], which are used independently of the nature of the solution. The problem is that the first terms of the polynomial are strongly dependent on the nature of the system and consequently, in most cases these expressions are not thermodynamically consistent, as they do not satisfy the limiting behavior required by the thermodynamics of solutions. Therefore, the subsequent use of the equation $a_1(m_2,m_3)$

ABSTRACT

An analytical thermodynamically self-consistent equation is proposed for the dependence on composition of the solvent activity of ternary solutions with electrolytic and molecular components (polyols, sugars, etc.). This equation was derived on the basis of the sum of contributions from short and long range interactions to the excess Gibbs free energy of solution. The correlation of experimental data corresponding to 20 aqueous ternary systems was successfully carried out. The results obtained demonstrate an excellent fitting capability of the proposed equation.

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for the evaluation of other thermodynamic properties (activity coefficients of the solutes, etc.) will lead to erroneous results.

In this context and due to the lack of reliable analytical expressions to describe the dependence $a_1(m_2, m_3)$, the aim of the present work is to develop a thermodynamically consistent simple multiparametric equation that can represent the experimental dependence of the solvent activity on composition with high accuracy for ternary solutions constituted by one electrolytic compound and one molecular compound in the whole range of composition. Moreover, the resulting expression will be suitable for the application of the McKay-Perrin [12,13], Canagaratna [14] or de Pablo [15] methods for the calculation of the activity coefficients of the other components of the solution, as well as for any other calculations of thermodynamic properties.

2. Theoretical considerations

2.1. Multiparametric equation

It is considered a ternary solution constituted by a solvent (1) an electrolyte (2) and a molecular component (3). The electrolyte its characterized by the formula unit $A_{\nu_A}C_{\nu_C}$,

$$A_{\nu_A}C_{\nu_C} \to \nu_A A^{Z_A} + \nu_C C^{Z_C} \tag{1}$$

Being $v_A(v_C)$ the stoichiometric coefficient of the anion (cation) corresponding to the dissociation of the electrolyte (2). The molality (m_i) is the concentration used for both components, however in order to develop the multiparametric equation of the solvent activity $a_1(m_2, m_3)$, the independent variables $z = m_2^{1/2}$ and

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 m_3 are adopted. The first one is chosen in order to make compatible the behavior of the solution at infinite dilution ($m_i \rightarrow 0$) with the way in which the correlation equation is set. In this context, the dependence $a_1(z,m_3)$ can be described by a Mclaurin series expansion as follows,

$$a_{1}(z, m_{3}) = 1 +A_{11}z + A_{12}m_{3} +A_{21}z^{2} + A_{22}zm_{3} + A_{23}m_{3}^{2} +A_{31}z^{3} + A_{32}z^{2}m_{3} + A_{33}zm_{3}^{2} + A_{34}m_{3}^{3} +A_{41}z^{4} + A_{42}z^{3}m_{3} + A_{43}z^{2}m_{3}^{2} + A_{44}zm_{3}^{3} + A_{45}m_{3}^{4} +A_{51}z^{5} + \dots$$

$$(2)$$

This expression contains 15 coefficients, being many of them determined by the limiting behavior of this type of solutions, which is developed in the following item.

2.2. Analytical description of the behavior of dilute solutions

The coefficients A_{ij} of the expression $a_1(z, m_3)$ given by Eq. (2) must be consistent with the constrains imposed by the Thermodynamics of solutions. In order to ensure this consistency, a condition of third order contact with the limiting behavior is applied to the proposed expression. Consequently, an exact dependence of $a_1(z, m_3)$ for diluted solutions is required. This can be obtained from the integration of the Gibbs–Dühem equation on terms of activity coefficients. The equation at constant temperature and neglecting the effect of pressure change with composition can be written as,

$$n_1^o d\ln\gamma_1 + \nu m_2 d\ln\gamma_+^o + m_3 d\ln\gamma_3^o = 0 \tag{3}$$

The corresponding reference states for the activity coefficients are perfect solution for the solvent (γ_1) and ideal dilute solution for components 2 (γ_{\pm}^o) and 3 (γ_3^o). From the integration of Eq. (3) along constant $x_3^o = m_3/(\nu m_2 + m_3)$, the following expression for the logarithm of the activity coefficient of solvent is obtained (see derivation in Appendix),

$$\ln \gamma_1 = \frac{A^{\gamma}}{3\sqrt{2}n_1^o} (\nu_A z_A^2 + \nu_C z_C^2)^{3/2} m_2^{3/2} + \frac{Bm_3^2 + 0.5(B - C)\nu m_2 m_3}{(n_1^o + \nu m_2 + m_3)^2}$$
(4)

Thus, the solvent activity of dilute solutions constituted by an electrolyte and a molecular solute can be described,

$$\ln a_{1} = \ln \left(\frac{n_{1}^{o}}{n_{1}^{o} + \nu m_{2} + m_{3}} \right) + \frac{A^{\nu}}{3\sqrt{2}n_{1}^{o}} (\nu_{A}z_{A}^{2} + \nu_{C}z_{C}^{2})^{3/2}m_{2}^{3/2} + \frac{Bm_{3}^{2} + 0.5(B - C)\nu m_{2}m_{3}}{(n_{1}^{o} + \nu m_{2} + m_{3})^{2}}$$
(5)

where the first term on the right is the logarithm of the solvent molar fraction. Eq. (5) describes the asymptotic behavior that must follow $a_1(m_2, m_3)$.

Coefficients A_{ij} can be related to the limiting experimental behavior starting from Eq. (5). Differentiating Eq. (5) with respect to z,

$$\left(\frac{1}{a_1}\frac{\partial a_1}{\partial z}\right) = -\frac{2\nu_2 z}{(n_1^o + \nu_2 z^2 + m_3)} + 4A^{\gamma}\nu_2 z n_1^o \frac{(\nu_2 z^2 + m_3)}{(n_1^o + \nu_2 z^2 + m_3)^3}$$
(7)

and taking the limit for the solvent molar fraction $x_1 \rightarrow 1$, the coefficient A_{11} is evaluated,

$$A_{11} = \left(\lim_{x_1 \to 1} \frac{\partial a_1}{\partial z}\right) = 0 \tag{8}$$

Then the differentiation with respect to m_3 ,

$$\left(\frac{1\,\partial a_1}{a_1\,\partial m}\right) = -\frac{1}{\left(n_1^0 + \nu_2 z^2 + m_3\right)} + 2A^{\gamma} n_1^0 \frac{\left(\nu_2 z^2 + m_3\right)}{\left(n_1^0 + \nu_2 z^2 + m_3\right)^3} \tag{11}$$

leads to coefficient A_{12} ,

$$A_{12} = \left(\lim_{x_1 \to 1} \frac{\partial a_1}{\partial m_3}\right) = -\frac{1}{n_1^o}$$
(12)

Applying the same procedure with the derivatives of higher order of Eq. (5) and taking into account the relationship between the derivatives of $\ln a_1$ and those of a_1 , it can be demonstrated the following identities,

$$A_{21} = \left(\frac{1}{2x_1 \to 1} \frac{\partial^2 a_1}{\partial z^2}\right) = -\frac{\nu}{n_1^o}$$
(13)

$$A_{22} = \left(\lim_{x_1 \to 1} \frac{\partial^2 a_1}{\partial m_3 \partial z}\right) = 0 \tag{14}$$

$$A_{23} = \left(\frac{1}{2}\lim_{x_1 \to 1} \frac{\partial^2 a_1}{\partial m_3 2}\right) = \frac{(1+2B)}{2n_1^0 2}$$
(15)

$$A_{31} = \left(\frac{1}{6} \lim_{x_1 \to 1} \frac{\partial^3 a_1}{\partial z^3}\right) = \frac{A^{\gamma}}{3\sqrt{2n_1^o}} \left(\nu_A z_A^2 + \nu_C z_C^2\right)^{3/2}$$
(16)

$$A_{32} = \left(\frac{1}{2}\lim_{x_1 \to 1} \frac{\partial^3 a_1}{\partial z^2 m_3}\right) = \frac{\nu(4+B-C)}{2n_1^0 2}$$
(17)

$$A_{33} = \left(\frac{1}{2}\lim_{x_1 \to 1} \frac{\partial^3 a_1}{\partial z m_3^2}\right) = 0 \tag{18}$$

$$A_{34} = \left(\frac{1}{6}\lim_{x_1 \to 1} \frac{\partial^3 a_1}{\partial m_3^3}\right) = -\frac{(3+18B)}{6n_1^0 3}$$
(19)

Substituting the coefficients given in Eqs. (8,12-19) into Eq. (2), the following expression of the dependence $a_1(z, m_3)$ is obtained,

$$a_{1}(z, m_{3}) = 1 - \frac{1}{n_{1}^{0}}m_{3} - \frac{\nu}{n_{1}^{2}}z^{2} + \frac{(1+2B)}{2n_{1}^{0}2}m_{3}^{2} + \frac{A^{\gamma}}{3\sqrt{2}n_{1}^{0}} \Big[\left(\nu_{A}z_{A}^{2} + \nu_{C}z_{C}^{2}\right)^{3/2} \Big] z^{3} + \frac{\nu(4+B-C)}{2n_{1}^{0}2}z^{2}m_{3} - \frac{(3+18B)}{6n_{1}^{0}3}m_{3}^{3} + A_{41}z^{4} + A_{42^{2^{3}}}m_{3} + A_{43}z^{2}m_{3}^{2} + A_{44}z^{42}m_{3}^{3} + A_{45}m_{3}^{4} + A_{51}z^{5} + \dots$$
(20)

From the comparison of Eqs. (20) and (2), it can be observed that the number of coefficients is decreased from 15 to 8. Renaming the coefficients and expressing Eq. (20) on terms of total molality m_2 , the final expression is,

Table 1 Values of the param	Table 1 Values of the parameters of Eq. (21) evaluated from the correlation of N experimental points of ternary aqueous solutions of electrolytic (2) and molecular (3) compounds. Maximum total molality ($m_{i,\max}$), standard deviation (5)	the correlati	on of N expe	rimental poin	its of ternary i	aqueous solutic	ons of electrolytic	(2) and molecul	ar (3) compou	inds. N	laximum 1	total mola	ılity (m _{i,max}), stanı	lard deviation (S).
Molecular compound (3)	Electrolytic compound (2)	В	J	10 ² a	$10^{4} b$	$10^5 c$	10 ⁶ d	10 ⁷ e	f	N	m _{2,max}	m _{3,max}	S	Refs.
Xylose	Na benzenesulfonate	-0.9930	8.606	-1.765	14.33	1.008	3.789	-6.495	0.006518	79	2.50	3.42	0.00023	[17] [18]
Urea	Na benzenesulfonate	2.011	-4.026	-1.741	-5.031	-23.40	-6.613	-29.27	0.006357	92	2.50	4.12	0.00025	[17] [18]
Glycine	$MgSO_4$	4.013	95.53	-19.94	134.3	7556	-41180	930.3	0.1028	40	1.00	1.16	0.00046	[19]
D-Alanine	$MgSO_4$	0.2107	45.55	-20.70	195.4	-494.3	5085	-1640	0.1118	38	0.95	1.17	0.00043	[19]
L-Valine	$MgSO_4$	-1.923	34.32	-26.96	-290.7	24170	-217200	-19592.7	0.2082	32	0.37	0.39	0.00006	[19]
Sorbitol	NaCI	-0.7167	4.665	-1.199	10.11	50.73	-851.7	-103.4	0.002250	62	6.00	3.72	0.00102	[20] [21]
β-Alanine	Tetra <i>n</i> -propylammonium Br	-0.9075	-6.122	-0.8828	-29.54	-341.4	309.8	-79.12	0.001384	98	6.33	6.88	0,00132	[22]
Sucrose	NaCl	-6.061	3.187	-1.210	14.60	-1.298	21.98	-4.124	0.002296	151	6.14	6.09	0.00122	[21] [23] [24]
Sorbitol	NaCl	-1.480	1.165	-1.253	6.509	-3.877	2.233	-5.746	0.002518	95	5.23	11.92	0.00076	[21] [25]
Sorbitol	KCI	-1.484	18.43	-1.247	-38.50	1046	-4073	-5.697	0.003147	56	3.82	11.92	0.00076	[21] [25]
L-Serine	Di-ammonium citrate	4.479	1.423	-9.330	6.403	-103.8	26.07	-238.4	0.03720	50	1.68	3.99	0.00050	[26]
Glycine	Di-ammonium citrate	3.548	-4.414	-10.72	-7.589	-168.3	162.0	-479.5	0.05094	35	1.16	2.55	0.00037	[26]
L-Alanine	Di-ammonium citrate	-0.5274	1.750	-12.08	37.89	-195.2	728.5	185.1	0.06700	35	0.88	1.75	0.00023	[26]
Xylose	NaCl	-0.6631	11.73	-1.446	11.91	72.04	-166.7	-88.73	0.003638	4	3.49	3.47	0.00077	[21] [27] [28]
Fructose	NaCl	-2.073	-0.8692	-1.481	1.355	35.93	-60.46	-56.32	0.003844	55	3.52	7.23	0.00125	[21] [28] [29]
Glucose	NaCI	-1.795	4.663	-1.366	6.485	18.45	-20.85	-6.147	0.003147	77	4.00	6.20	0.00097	[21] [28] [30]
Glycine	KCI	3.808	-12.01	-1.459	-36.14	-142.3	-2.450	-392.4	0.004700	49	1.62	3.32	0.00024	[31]
Urea	Na ₂ SO ₄	1.6377	12.0845	-7.4204	10.7668	7.9760	2.6561	5.4204	0.02217	82	3.00	9.97	0.00151	[32] [33]
Urea	NaCl	1.706	5.540	-1.190	2.854	-0.7656	0.5195	3.984	0.002223	83	6.21	10.00	0.00073	[34]
Fructose	KCI	-8.632	-21.825	-1.1054	-18.1157	-60.789	330.404	3565.48	0.00236	48	4.25	2.43	0.00073	[35]

$$a_{1}(m_{2},m_{3}) = 1 - \frac{1}{n_{1}^{0}} \left\{ m_{3} + \nu m_{2} - \frac{A^{\gamma}}{3\sqrt{2}} [(\nu_{A}z_{A}^{2} + \nu_{C}z_{C}^{2})^{3/2}]m_{2}^{3/2} \right\} + \frac{(1+2B)}{2n_{1}^{0}2}m_{3}^{2} + \frac{\nu(4+B-C)}{2n_{1}^{0}2}m_{2}m_{3} - \frac{(3+18B)}{6n_{1}^{0}3}m_{3}^{3} + am_{2}^{2} + bm_{2}^{3/2}m_{3} + cm_{2}m_{3}^{2} + dm_{2}^{1/2}m_{3}^{3} + em_{3}^{4} + fm_{2}^{5/2} + \dots$$
(21)

Eq. (21) is a thermodynamically consistent expression that can accurately correlate the experimental dependence $a_1(m_2,m_3)$ in the whole concentration range for ternary solutions of an electrolyte and a molecular component, as it will be verified through its application to several systems, which will be described in the next section.

3. Evaluation of the proposed equation

The proposed equation ensures that the description of the dependence $a_1(m_2, m_3)$ has at the origin a third order contact with the limiting behavior established for this type of solution. Thus, it is possible to describe the limiting behavior of solvent activity transition between electrolyte and molecular component by changing x_3^0 . For instance when $m_3 = 0$ ($x_3^0 = 0$), Eq. (21) turned to be equal to the expression for ternary electrolyte solutions, which was previously derived through a different approach [16], when the internal molal fraction y is 0 or 1 (binary system).

In order to verify the applicability of Eq. (21,20) aqueous ternary systems were evaluated, constituted by electrolytes of the type 1:1. 1:2, 2:2 and molecular components as polyols, sugars, etc. Eight adjustable parameters were used, as it was realized that they were enough for the appropriate correlation (a_1^{corr}) of the experimental dependence (a_1^{exp}) in the whole concentration range. All the evaluated systems were at 298.15 K, thus A^{γ} = 1.19295, being the solvent molality $n_1^o = 55.5084$.

The results obtained are summarized in Table 1, where it is reported for each system the values of the eight coefficients of Eq. (21), the number of experimental data (N) involved in the correlation, the maximum concentration of the solutes $(m_{i,max})$, the standard deviation $S = (\sum (a_1^{\text{corr}} - a_1^{\text{exp}})^2)^{1/2}$ and the references from where the experimental data were obtained. The correlation capability of the proposed equation is illustrated for four cases that can be considered representative of all the systems analysed. Figs. 1–4 show the dependencies $a_1(m_2, m_3)$ for different systems, involving very different types of electrolytes with different molecular compounds, through which the capability of Eq. (21) to describe the effect of the variation of the ionic charges is verified. Furthermore, from the analysis of the S column of Table 1, it can be concluded that the standard deviations of the 20 systems analyzed are between $6 \times 10^{-5} < S < 1.5 \times 10^{-3}$, observing an average value of S equal to 0.000693, which is very low, even when it is compared with those obtained previously for ternary electrolyte systems [16]. It can be also favorably compared with another much more complex equation, with a larger number of adjustable parameters [36]. Taking into account that the values of the standard deviation include errors that might have occurred during the experimental measurements, it can be concluded that the proposed equation, which is consistent with the limiting behavior demanded by the Thermodynamics of solutions, is an accurate tool for the description of the dependence of the solvent activity on composition of ternary solutions with electrolyte and molecular components.

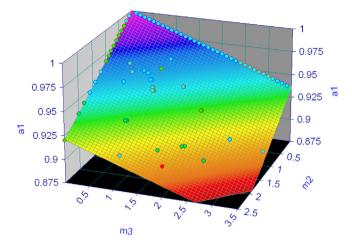


Fig. 1. Dependences of solvent activity (a_1) on molality of both components for the system sodium bencenesulfonate (2) – xylose (3) – water(1) at 298.15 K. Dots: experimental data [17,18]; surface: correlation with Eq. (21).

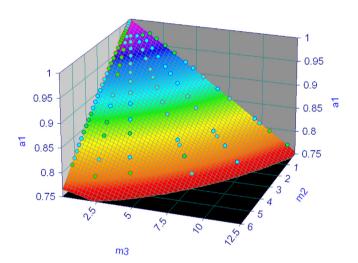


Fig. 2. Dependences of solvent activity (a_1) on molality of both components for the system sodium chloride (2) – sorbitol – water (1) at 298.15 K. Dots: experimental data [20,21]; surface: correlation with Eq. (21).

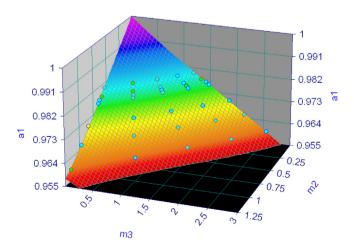


Fig. 3. Dependences of solvent activity (a_1) on molality of both components for the system di-ammonium citrate (2) – glycine – water (1) at 298.15 K. Dots: experimental data [26]; surface: correlation with Eq. (21).

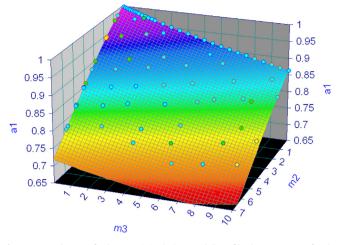


Fig. 4. Dependences of solvent activity (a_1) on molality of both components for the system sodium chloride (2) – urea – water (1) at 298.15 K. Dots: experimental data [34]; surface: correlation with Eq. (21).

4. Conclusion

Starting from a Mclaurin series expansion of the solvent activity in ternary electrolyte (2) and molecular (3) component solutions on the variables m_2 and m_3 and taking into account the limiting behavior that must be accomplished, an analytical equation was derived that allows the correlation of the dependence of the solvent activity on composition. This equation was applied to 20 ternary systems and the results obtained demonstrate an excellent fitting capability of the experimental data. The main advantage of the proposed equation is the direct correlation of the solvent activity, which avoids errors introduced by indirect methods, especially in the region of dilute solutions.

Appendix.

Derivation of Eq. (4)

For the derivation of the dependence of the logarithm of the activity coefficient of the solvent on concentration in dilute solutions, the integration of Gibbs–Dühem Eq. (3) needs the expressions of the logarithm of the activity coefficients of the other components. These expressions can be obtained through the application of the criterion of additivity of the contributions of short (*s*) and long (*l*) range interactions,

$$\ln \gamma_i = \ln \gamma_i^s + \ln \gamma_i^l \tag{A1}$$

Taking into account that it is intended to describe the behavior at infinite dilution, $x_1 \rightarrow 1$, it can be considered only one term of Eq. (A1). For the electrolyte (2), the short range interaction contribution can be neglected, being the long range interaction given by the Debye–Huckel equation,

$$\ln \gamma_{\pm}^{o} = -\frac{A^{\gamma}}{3\sqrt{2}n_{1}^{o}} z_{+}|z_{-}|(\nu_{A}z_{A}^{2} + \nu_{C}z_{C}^{2})^{1/2}m_{2}^{1/2}.$$
 (A2)

On the other hand, for the molecular solute (3) the long range interaction contribution can be neglected and the short range interaction can be described by the Porter equation for a ternary solution [37,38],

$$\ln\gamma_3 = a_{13}x_1^2 + a_{23}x_2^2 + x_1x_2(a_{13} + a_{23} - a_{12}) \tag{A3}$$

where x_i (*i*:1,2,3) is the molar fraction of the components. The activity coefficient needed is that corresponding to the infinite

dilution solution reference state, $\ln \gamma_3^o$, which can be calculated from Eq. (A3) as,

$$\ln\gamma_3^o = \ln\gamma_3 - \underset{x_1 \to 1}{\lim} \ln\gamma_3 \tag{A4}$$

Considering that the second term of Eq. (A3) can be neglected for dilute solutions, the final expression for $\ln \gamma_3^o$ can be written as,

$$\ln \gamma_3^{\rm o} = \mathbf{B}(x_1^2 - 1) + x_1 x_2 (\mathbf{B} + \mathbf{C}) \tag{A5}$$

where $B = a_{13}$ and $C = a_{23} - a_{12}$. Furthermore, the internal molal fraction is,

$$x_i^o = \frac{\nu_i m_i}{(\nu m_2 + m_3)}$$
 i:2,3 $\nu_3 = 1$ $\nu_2 = \nu_A + \nu_C$ (A6)

being the relationship with the solvent molar fraction x_1 ,

$$x_i = x_i^0 (1 - x_1)$$
 (A7)

Substituting Eqs. (A2) and (A5) into Eq. (3), holding x_i^o constant and integrating between the limits $x_1 = x_1$ and $x_1 = 1$, the following expression is obtained,

$$\begin{split} &\ln \gamma_1 = -\frac{A^{\gamma}}{3\sqrt{2}n_1^o} (\nu_A z_A^2 + \nu_C z_C^2)^{3/2} m_2^{3/2} + x_3^o \Big(B - x_2^o (B + C)\Big) (1 - x_1)^2 \\ &- x_2^o x_3^o (B + C) \ln x_1 - x_2^o x_3^o (B + C) (1 - x_1) \end{split}$$
 (A8)

Eq. (A8) can be simplified taking the first three terms of the Taylor series expansion for $\ln x_1$, arriving finally to Eq. (4),

$$\ln\gamma_{1} = \frac{A^{\gamma}}{3\sqrt{2}n_{1}^{o}}(\nu_{A}z_{A}^{2} + \nu_{C}z_{C}^{2})^{3/2}m_{2}^{3/2} + \frac{Bm_{3}^{2} + 0.5(B - C)\nu m_{2}m_{3}}{(n_{1}^{o} + \nu m_{2} + m_{3})^{2}}$$
(4)

References

- [1] J.F. Comesaña, A. Correa, A.M. Sereno, J. Chem. Eng. Data 44 (1999) 1132–1134.
- [2] D. Lin, Z. Zhu, L. Mei, L. Yang, J. Chem. Eng. Data 41 (1996) 1040-1042.
- [3] F. Deumier, P. Bohuon, J. Food Eng. 68 (2005) 377–383.
- [4] M. Zafarani-Moattar, S. Sarmad, J. Chem. Eng. Data 55 (2010) 5182–5190.
- [5] M. Zafarani-Moattar, S. Sarmad, J. Chem. Eng. Data 55 (2010) 5182–5190.
 [6] Y. Liu, C. Xu, Y. Hu, J. Yan, Chin. J. Chem. Eng. 12 (2004) 470.
- [7] S.A. Cooke, S.O. Jonsdottir, P. Westh, J. Chem. Thermodyn. 34 (2002) 1545–1555.
- [8] M.D. Cohen, R.C. Flagan, J.H. Seinfeld, J. Phys. Chem. 91 (1987) 4563-4574.
- [9] A. Eleassi, H. Modarress, J. Chem. Eng. Data 44 (1999) 52-56.
- [10] T.T. Teng, F. Lenzi, J. Can. Chem. Eng. 52 (1974) 387-391.
- [11] A. Dinane, Fluid Phase Equilib. 273 (2008) 59–76.
- [12] H.A.C. McKay, J.K. Perring, Trans. Faraday Soc. 49 (1953) 163-165.
- [13] H.A.C. McKay, Trans. Faraday Soc. 49 (1953) 237–242.
 [14] G. Canagaratna, M. Maheswaran, J. Chem. Soc. Trans. Faraday 80 (1984)
- 261–265. [15] A. Kolker, J.J. de Pablo, Chem. Eng. Sci. 50 (1995) 1953–1955.
- [16] A. Ricardini, F. Passamonti, A.C. Chialvo, Fluid Phase Equilib. 345 (2013) 23–27.
- [17] O.D. Bonner, O.C. Rogers, J. Phys. Chem. 64 (1960) 1499-1501.
- [18] H. Uedaira, H. Uedaira, J. Phys. Chem. 74 (1970) 1931-1936.
- [19] A. Salabat, S. Neshat, A. Fazlali, Fluid Phase Equilib. 314 (2012) 198-202.
- [20] V.E. Bower, R.A. Robinson, J. Phys. Chem. 67 (1963) 1540-1541.
- [21] R.A. Robinson, R.H. Stokes, Electrolyte Solutions, fifth ed., Butterworth, London, 1970.
- [22] C.N. Pepela, P.J. Dunlop, J. Chem. Thermodyn. 4 (1972) 115-121.
- [23] R.A. Robinson, R.H. Stokes, J. Phys. Chem. 65 (1961) 1954–1958.
- [24] R.A. Robinson, R.H. Stokes, K.N. Marsh, J. Chem. Thermodyn. 2 (1970) 745–750.
- [25] O.D. Bonner, J. Solution Chem. 11 (1982) 315–324.
- [26] R. Sadeghi, A. Gholamireza, J. Chem. Thermodyn. 43 (2011) 200–215.
- [27] H. Uedaira, H. Uedaira, Chem. Soc. Jpn. 42 (1969) 2137-2140.
- [28] J.F. Comesaña, A. Correa, A.M. Sereno, J. Chem. Eng. Data 44 (1999) 1132–1134.
- [29] A. Correa, J.F. Comesaña, A.M. Sereno, Int. J. Food Sci. Technol. 29 (1994)
- 331–338.
 [30] K. Miyajima, M. Sawada, M. Nakagari, Bull. Chem. Soc. Jpn. 56 (1983) 1620–1623.
- [31] V.E. Bower, R.A. Robinson, J. Res. Natl. Bur. Stand. Sec. A Phys. Chem. 69 (1965) 131–135.
- [32] E.E. Schrier, R.A. Robinson, J. Biol. Chem. 245 (1970) 2432–2437.
- [33] M. El Guendouzi, A. Mounir, E. Dinane, J. Chem. Thermodyn. 35 (2003) 209–220.
- [34] V.E. Bower, R.A. Robinson, J. Phys. Chem. 67 (1963) 1524-1527.
- [35] A. Derossi, C. Severini, Eur. Food Technol. 223 (2006) 216-224.
- [36] Y.F. Hu, T.M. Guo, Phys. Chem. Chem. Phys. 1 (1999) 3303-3308.
- [37] A.W. Porter, Trans. Faraday Soc. 16 (1921) 336–345.
- [38] J.P. O'Connell, J.H. Haile, Thermodynamics: Fundamentals for Applications, Cambridge Univ. Press, New York, 2005, pp. 216.