

# **ELECTROLYTES: EQUILIBRIA IN SOLUTIONS AND PHASE EQUILIBRIA. CALCULATION OF MULTICOMPONENT SYSTEMS AND EXPERIMENTAL DATA ON THE ACTIVITIES OF WATER, VAPOR PRESSURES, AND OSMOTIC COEFFICIENTS**

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# **Introduction**

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The edition is a continuation of a series of works of this author that are devoted to many-years' theoretical studies of the structure of, and interparticle interactions in, aqueous solutions of electrolytes, programmed and factographic representation of experimental data, and recommendations related to the methods of calculating physicochemical parameters of binary and multicomponent systems [1–24].

In this edition, information on the vapor pressures over solutions, activities of water, and osmotic coefficients over electrolyte solutions is substantially remade in comparison with [19, 21], and new experimental data are given. Coefficients for calculating vapor pressures over solutions of a large number of electrolytes that were obtained by mathematical treatment (using methods of regression analysis) of experimental data available in the literature are presented. Experimental data for most widely applied electrolytes are given for a high-temperature range.

In Part I, methods for calculating vapor pressures over solutions, activities of water, osmotic coefficients, and boiling and freezing temperatures of multicomponent solutions of electrolytes are given; related equations for calculating water vapor pressures over pure water are considered.

In Part II, the list of electrolytes for which vapor pressures, activities of water, and osmotic coefficients over solutions were calculated is extended in comparison with [21] (see List of tables to Part II); the existing tables were remade. In this issue, as in [21], a logical mode of representing reference data was retained; for each electrolyte, the maximum temperature range and the maximum range of concentrations are given. A large attention is paid to high-temperature researches. For many electrolytes, additional original experimental investigations were carried out. The author hopes that all these changes will favor the readability and applicability of the material presented.

In this edition, the following system of references is used. For each property and each electrolyte, the full bibliography is given (although many data were refined by our own experimental studies, and it is by no means always that information from the references mentioned has been mathematically processed); in those cases where no references are given, this means that original data of this author were used.

The author will be very grateful to readers for any remarks and wishes.

# Part I

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## 1 Multicomponent Solutions of Electrolytes. Calculation of Equilibria in Solutions and Phase Equilibria

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### 1.1 Vapor Pressures and Activities of Water

Activities of water can be calculated using the thermodynamic equation

$$\alpha_w = p/p_0 \quad (1.1)$$

where  $p$  is the vapor pressure (Pa) of water over a solution and  $p_0$  is the vapor pressure (Pa) of water over pure water.

The vapor pressure of water over pure water is calculated with a root-mean-square error of 39.54 J/(kg · K) and mean relative error of 0.006% using the formula

$$p_0 = P_{\text{cr}} \exp B \quad (1.2)$$

where  $P_{\text{cr}}$  is the critical pressure (equal to  $2.2064 \cdot 10^7$  Pa) and  $B$  is defined as

$$B = T_{\text{cr}} A/T \quad (1.3)$$

where  $T_{\text{cr}}$  is the critical temperature (647.14 K),  $T$  is temperature (in K), and  $A$  is determined from the following formulas:

from 0 to 100°C:

$$A = A_0 + A_1 T_*^{1.5} + A_2 T_*^3 + A_3 T_*^{3.5} \quad (1.4)$$

where  $A_0 = -0.595684$ ,  $A_1 = -11.039345$ ,  $A_2 = 17.449275$ , and  $A_3 = -16.028445$ ;

from 100 to 250°C:

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$$A = A_0 + A_1 T_* + A_2 T_*^{1.5} + A_3 T_*^4 + A_4 T_*^{4.5} \quad (1.5)$$

where  $A_0 = -0.018527$ ,  $A_1 = -7.284175$ ,  $A_2 = 0.407838$ ,  $A_3 = -4.200201$ , and  $A_4 = 1.005549$ ;

from 250 to 350°C:

$$A = A_0 + A_1 T_* + A_2 T_*^{1.5} + A_3 T_*^3 + A_4 T_*^{3.5} + A_5 T_*^4 \quad (1.6)$$

where  $A_0 = -0.001096$ ,  $A_1 = -7.891114$ ,  $A_2 = 2.024116$ ,  $A_3 = -22.917717$ ,  $A_4 = 60.32581$ , and  $A_5 = -50.818889$ .

In Eqs. (1.4)–(1.6), the temperature  $T_*$  is defined as

$$T_* = 1 - T/T_{\text{cr}} \quad (1.7)$$

The vapor pressure of water over an electrolyte solution is calculated by the equation

$$p = p_0 \exp \left( 2.3026 \sum_{i=1}^k m_i P_i^* \right) \quad (1.8)$$

where  $m_i$  is the electrolyte concentration in a solution (mol/kg H<sub>2</sub>O);  $k$  is the number of components in the solution; and  $P_i^*$  is calculated by the formula

$$P_i^* = W_{0i} + W_{1i}t_i + W_{2i}m_i + W_{3i}t_i^2 + W_{4i}t_i m_i + W_{5i}t_i^2 m_i \quad (1.9)$$

where the coefficients  $W_{ji}$  are given for a number of electrolytes in Table 1.1.

Other methods of calculating activities of water are considered in [3, 4, 10, 11, 15, 18, 20, 21, 24]. Experimental data for the activities of water and vapor pressures of water over electrolyte solutions in wide ranges of temperature and solute concentrations are given in Part II.

**Example.** Calculate the vapor pressure and the activity of water over a CaCl<sub>2</sub>–NaCl–H<sub>2</sub>O solution at  $T = 50^\circ\text{C}$  and solute concentrations 10 wt. % CaCl<sub>2</sub> and 5 wt. % NaCl.

The experimental vapor pressure over the solution is  $p = 11100 \text{ Pa}$  [217]. The molecular weights are  $M_{\text{CaCl}_2} = 110.986$  and  $M_{\text{NaCl}} = 58.443$ . The absolute temperature is  $T = 273.15 + 50 = 323.15 \text{ K}$ .

Using (1.7), we calculate  $T_*$ :

$$T_* = 1 - 323.15/647.14 = 0.500649$$

Using (1.4), we calculate  $A$ :

$$\begin{aligned} A &= -0.595684 - 11.039345 \cdot 0.500649^{1.5} + 17.449275 \cdot 0.500649^4 \\ &\quad - 16.028445 \cdot 0.500649^{3.5} = -3.739794. \end{aligned}$$

Using (1.3), we calculate  $B$ :

$$B = 647.14(-3.739794)/323.15 = -7.48931$$

Using (1.2), we find the vapor pressure of water over pure water:

$$p_0 = 2.2064 \cdot 10^7 \exp(-7.48931) = 12334.4 \text{ Pa}$$

In order to calculate the vapor pressure of water over the solution using (1.8), we preliminarily calculate the concentration of the solution components in moles per kg of H<sub>2</sub>O:  
for CaCl<sub>2</sub>:

$$m_i = \frac{1000 c_i}{M_i \left( 100 - \sum_{i=1}^k c_i \right)} = \frac{1000 \cdot 10}{110.986(100 - 10 - 5)} = 1.0600 \text{ mol/kg}$$

for NaCl:

$$m_j = \frac{1000 \cdot 5}{58.443(100 - 10 - 5)} = 1.0065 \text{ mol/kg}$$

Select coefficients  $W_{ji}$  for each electrolyte:

for CaCl<sub>2</sub>:

$$\begin{array}{lll} W_{0i} = -0.020638 & W_{1i} = 0.0001245 & W_{2i} = -0.0083089 \\ W_{3i} = -0.0000007 & W_{4i} = 0.0000006 & W_{5i} = 0.0000002 \end{array}$$

for NaCl:

$$\begin{array}{lll} W_{0i} = -0.01505 & W_{1i} = -0.0000508 & W_{2i} = -0.0008033 \\ W_{3i} = 0.0000004 & W_{4i} = 0.0000068 & W_{5i} = -0.000000034 \end{array}$$

Calculate  $P_i^*$  by (1.9):

for CaCl<sub>2</sub>:

$$\begin{aligned} P_1^* &= -0.020638 + 0.0001245 \cdot 50 - 0.0083089 \cdot 1.0600 - 0.0000007 \cdot 50^2 \\ &\quad + 0.000006 \cdot 50 \cdot 1.0600 + 0.0000002 \cdot 50^2 \cdot 1.0600 = -2.412256 \cdot 10^{-2} \end{aligned}$$

for NaCl:

$$\begin{aligned} P_2^* &= -0.01505 - 0.0000508 \cdot 50 - 0.0008033 \cdot 1.0065 - 0.0000004 \cdot 50^2 \\ &\quad + 0.0000068 \cdot 50 \cdot 1.0065 - 0.000000034 \cdot 50^2 \cdot 1.0065 = -1.714187 \cdot 10^{-2} \end{aligned}$$

The vapor pressure of water over the electrolyte solution is calculated by (1.8):

$$\begin{aligned} p &= 12334.4 \exp\{2.3026[1.0600(-2.412256 \cdot 10^{-2}) \\ &\quad + 1.0065(-1.714187 \cdot 10^{-2})]\} = 11176 \text{ Pa (0.68\%)} \end{aligned}$$

The activity of water is determined by (1.1):

$$a_w = 11176 / 12334.4 = 0.9061$$

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