ELECTROLYTES. INTERPARTICLE INTERACTIONS. THEORY, CALCULATION METHODS, AND EXPERIMENTAL DATA

ELECTROLYTES. INTERPARTICLE INTERACTIONS. THEORY, CALCULATION METHODS, AND EXPERIMENTAL DATA

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ELECTROLYTES. INTERPARTICLE INTERACTIONS. THEORY, CALCULATION METHODS, AND EXPERIMENTAL DATA

The mathematical foundations of a theory of ion-ion interactions in concentrated electrolyte solutions were given. The potential of ion-ion interaction, a theory of ionic forces, a thermodynamic theory of equilibrium in solutions, and a theory of transport phenomena were considered. The theoretical material was illustrated by many tables and calculation examples.

The theoretical, semi-empirical, and empirical methods of calculation of the physicochemical parameters of the activity of water, activity coefficients, a decrease in the vapor pressure over solutions, the electrical conductivity, viscosity, diffusivity, and some other parameters were analyzed. Calculation coefficients derived from a mathematical treatment of the available experimental data were given, and the errors for many tens of salts were presented. The material was illustrated by numerous calculation examples.

Tables of the experimental data for phase equilibria and equilibria in solutions (decrease of the vapor pressure over solutions, water activity, activity coefficients) and transport properties were presented in a wide temperature range and a wide range of mass contents of electrolytes in solutions. The experimental data were treated by computer.

The book contains the data for a wide range of electrolytes which are most often used in modern chemical technology. The book is intended for scientists and engineers working in chemical industry and allied industries.

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Foreword

The book is the continuation of a number of works of the author [1–3] dealing with investigation, representation, and methods of calculation of the physicochemical properties of binary and multicomponent electrolyte solutions.

A mathematical theory of concentrated electrolyte solutions is first given in such a complete form. The potential of ion-ion interaction, a theory of ionic forces, a thermodynamic theory of equilibria in solutions, a theory of transport phenomena are considered. Unfortunately, complex processes that occur in electrolyte solutions cannot virtually be described by simple mathematical procedures; therefore, an adequate background is required to understand many mathematical calculations. Each section of the theory is finished by an equation to describe any property and is illustrated by calculation examples.

The author will gratefully accept all comments and wishes of the reader.

MATHEMATICAL FOUNDATIONS OF A THEORY OF INTERPARTICLE INTERACTIONS IN CONCENTRATED ELECTROLYTE SOLUTIONS

1

General Characteristics of the Problem of Interparticle Interactions in Electrolyte Solutions

Nowadays, the Debye-Hückel theory [4], which is concerned with an order in the distribution of ions in solutions, is the most widely used classic electrostatic theory of dilute solutions, and its basic points are the concept of ionic atmosphere and its thickness. By the ionic atmosphere, P. Debye and E. Hückel meant that each ion is surrounded by a certain density of electric charges. The properties of solutions of strong electrolytes depend on the ratio of the electrostatic forces of ion fields, caused by their charges, and the properties of the medium, expressed by the dielectric constant of a solvent.

The Debye-Hückel theory correctly describes highly diluted electrolyte solutions. The further development of the electrostatic theory was due to description of more concentrated

solutions. One of the directions was the way of solving the equations of the Debye-Hückel theory as exactly as possible.

Hückel tried to extend the theory to the region of concentrated solutions, by assuming that the dielectric constant of water decreases in a solution and depends on the kind of ions. This attempt was failed, because the data on the effect of the ion charge on the dielectric constant and its concentration dependence were virtually unavailable. Moreover, experimental investigations showed that the dielectric constant increased with the electrolyte concentration.

Then, other scientists tried to take into account terms of a higher order in expansion of the hyperbolic sine into a series. Although a rather powerful mathematics apparatus was used, no significant progress in the region of high concentrations was achieved. The disadvantages of the Debye-Hückel theory may be the fact that it did not take into account ion association, solvating, complex formation, association of the hydrate shells of a solvent, etc.

In the last few decades, theories of electrolyte solutions given, for example, in the works [5–18] have gained wide acceptance. In these works, the Debye-Hückel theory was interpreted to some extent or some aspects of solvating, complex formation, association of the hydrate shells of a solvent, etc. were considered. A statistic quantum theory of solutions, which is rapidly developed owing to accumulation of modern experimental results and wide use of computers [17–19], has achieved some success, which allows one to obtain information on the state of particles. This information, however, is always tightly connected with the structure of a solvent.

1.1 Structural Properties of a Solvent

One of the basic problems of a theory of a liquid state is the problem of the structure of liquid because the structure of substance is tightly connected with its physical, chemical, and other properties. Currently, there is good reason to believe that the structure of liquid should be taken into account in solving the problems of a theory of liquid solutions and that further development of a theory of aqueous electrolyte solutions should involve investigation of their structures [20].

The main part of a solution is a solvent. Therefore, a theory of solutions describing a wide range of concentrations, temperatures, and solvents cannot be advanced without a knowledge of the structures of pure liquids used as solvents [21]. Two main aspects are usually highlighted in a theory of solutions: the structures of pure liquids (solvents) and the structures of solutions.

One of the most used solvents is water. Water is the most abundant liquid throughout the Earth, and it plays an important role in various processes that occur in animate and inanimate nature. Although the function of water as a solvent favoring many processes was discussed in a lot of papers, no common understanding of the structure of water was achieved. Numerous models of the structure of water, which explained various properties of water, were developed. The structure of water was studied in many works [5, 20, 22–24]. In these works, the following main features of a generalized model of liquid water may be seen: structural units and their relative position and mutual interaction. The structure of liquid water is characterized by a quasi-crystalline lattice, where a certain part of unit crystals is destroyed. The unit crystal of water consists of molecules of water or H^+ and O^{2-} ions. The relative position of the structural units of liquid water is such that they form

regions of an ordered icelike lattice with violation of the long-range order. The unit cell of the lattice is a tetrahedron with water molecules (OH^- or O^{2-} ions) in the center and vertices. The distance between the cores of oxygen is 2.9 Å from X-ray diffraction data and 2.86 Å from Raman spectra [24]. H^- ions have two quasi-stationary positions in a segment connecting oxygen cores. The distance of H^- ions from two ends of this segment. The interaction between the structural units of liquid water is due to hydrogen bond for all levels of the structure (single crystal, unit crystal, and water molecule) [25]. The hydrogen-bond energy for water is about 0.2 eV or 19 MJ/kmol.

A three-dimensional network of water molecules joint by extended and bent hydrogen bonds oscillates, which is accompanied by breaking and regeneration of hydrogen bonds. Each bond lives for some time and is broken for some time, which corresponds to 9% of broken bonds at 0°C and 20% of these at the boiling temperature [26].

Data on the basic thermodynamic properties of water are given in the works [27–30]. Precise experimental data on the dielectric constant of water are presented in [26].

1.2 The Structure of Electrolyte Solutions and Ionic Solvating

As solutions form, the structure of a solvent (water) undergoes changes caused by the interaction between electrolyte ions and the solvent. The interaction between ions and the solvent (water) is called ion solvating (hydration), and the number of molecules of water located in the immediate vicinity of an ion is called the coordination number of solvating (hydration). Strictly speaking, hydration is the transition of an ion from the standard state in the gas phase to the standard state in an aqueous solution. The phenomenon of solvating is complex and includes chemical and physical processes, and no common concept accounting for various aspects of this phenomenon has yet been achieved. L'vov and Zarembo [31] believe that solvating is a function of Coulomb, dipole, and dispersion interaction (pure ionic and covalent bonds usually have no place). The strength of ion-solvent bond is high (300–4000 kJ/mol and higher) [32].

Laidier and Muirhead-Gould [33] proposed two models of hydration of single-atom ions: discontinuous and continuous ones. The author concluded that the distribution of "bound" water around small ions is tetrahedral, and that around large ions is octahedral. In the continuous model of hydration, ion-dipole and ion-multidipole interactions are considered, and water polarization was not taken into account [34].

Celeda [35] believes that two solvent zones, made up of radially oriented molecules of a solvent and molecules with the normal structure of water, form around an ion.

In the model given in [36], a solvent plays the key role. The author analyzed the properties of solutions and the equilibrium between a zone with the "inherent structure" of water, where water remains in the state that it had prior to dissolving of an electrolyte, an intermediate zone, which is called a zone of "destructurized water", and a zone formed as a result of a direct effect of an ion, which is called "ion" zone. This point of view seems to be a more real concept for dilute solutions.

According to the points of view in [37], hydration of ions may be analyzed using the following two approaches: thermodynamic and kinetic ones. The former assumes the existence of a strong ion-water bond, whereas the latter considers the possibility of exchange reactions. The concept that the ion-water bond is saturated, which should be accepted, was grounded. This concept allows one to calculate the maximum number of

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