

# An Equation of State for Nonaqueous Electrolyte Solutions

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#### ABSTRACT

A two parameters equation of state (EOS) for nonaqueous electrolyte solutions system has been developed. The equation is in terms of Helmholtz free energy and incorporated with results of low density expansion of non-primitive mean spherical approximation. The EOS was tested for experimental data reported in literatures of 9 nonaqueous single electrolyte solutions of which the temperature was 298.15 K, and it also has a good predictive capability for nonaqueous electrolyte solutions at different temperature in this work. The comparisons with EOSs published earlier by other researchers in literatures are carried out in detail.

Keywords: EOS; Nonaqueous; Electrolyte Solutions

#### 1. Introduction

Electrolyte solutions are encountered in a wide variety of industrial processes, for example, wastewater treatment, extraction, seawater desalinization, distillation and geological processes. It is very important for us to describe the thermodynamic properties of such systems accurately. Phase equilibrium in electrolyte systems is very important to chemical industry.

Past a few decades, people made a lot of progress on describing thermodynamic properties of electrolyte systems with some models [1-4]. But most of studies in literatures were restricted in aqueous electrolyte systems. There is little attention on nonaqueous electrolyte systems until now, Although we can get some data of properties from literature, the data about nonaqueous electrolyte systems is much less than the one about aqueous electrolyte systems. So in engineering, we need a simple predictive model in order to describe phase behavior of nonaqueous electrolyte systems.

EOSs of nonaqueous electrolytes have been developed successfully since the late 1970s. Pitzer's models [5.6] have also extended to nonaqueous electrolyte solutions and the adjustable parameters are needed in all of these models. But up to now, there are still few models to represent phase equilibria properties of nonaqueous electrolyte solution.

In general, EOS can be derived by differentiating the Helmholtz free energy with respect to the density. The Helmholtz free energy is divided into several contributions, typically including selvent-solvent, ion-solvent and ion-ion terms. In this work, we expanded Helmholtz free energy as several contributions (including electrostatic contribution and association contribution) according to perturbation theory. On the other hand, the EOS proposed is tested for 9 nonaqueous solutions of alkali metal halides. The parameters can be obtained by fitting the vapor pressure of solvents. In addition, we also compared our results with the results of Mock et al., Youxiang Zuo and Tzuejen Chou.

#### 2. Model and Theory

The systems of interest in this work are limited in nonaqueous solutions (methanol solvent) of alkali metal halides. Since the salts are fully dissociated, there are three components in the solution, including cation, anion and methanol solvent respectively. The ions are treated as charged Lennard-Jones (LJ) spheres with additional associating sites forming hydrogen bonds with methanol. A methanol molecule is regarded as the LJ sphere with embedded a point dipole and three associating sites, two of which represent lone pair electrons and the others represent protons.

At temperature T and volume V, the system consists of N particles, and the number of species i is  $N_c$ . By using the perturbation theory [7-10], the differences of the Helmholtz free energies  $(A-A^{to})$  between the system and the corresponding hard sphere system can be expanded as

$$\frac{A - A^{bc}}{NkT} = \frac{A^{LF}}{NkT} + \frac{A^{obset}}{NkT} + \frac{A^{oos}}{NkT}$$
(1)

where k is the Boltzmann constant. The superscripts ks, LJ, elect and assoc represent the contributions from hard

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# **Table of Contents Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions**

- 1. Understanding the eBook Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - The Rise of Digital Reading Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Advantages of eBooks Over Traditional Books
- 2. Identifying Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Exploring Different Genres
  - Considering Fiction vs. Non-Fiction
  - Determining Your Reading Goals
- 3. Choosing the Right eBook Platform
  - Popular eBook Platforms
  - Features to Look for in an Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - User-Friendly Interface
- 4. Exploring eBook Recommendations from Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Personalized Recommendations
  - Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions User Reviews and Ratings

## **Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions**

- Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions and Bestseller Lists
- 5. Accessing Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions Free and Paid eBooks
  - Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions Public Domain eBooks
  - Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions eBook Subscription Services
  - Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions Budget-Friendly Options
- 6. Navigating Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions eBook Formats
  - o ePub, PDF, MOBI, and More
  - Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions Compatibility with Devices
  - Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions Enhanced eBook Features
- 7. Enhancing Your Reading Experience
  - Adjustable Fonts and Text Sizes of Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Highlighting and Note-Taking Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Interactive Elements Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
- 8. Staying Engaged with Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Joining Online Reading Communities
  - o Participating in Virtual Book Clubs
  - Following Authors and Publishers Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
- 9. Balancing eBooks and Physical Books Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Benefits of a Digital Library
  - Creating a Diverse Reading Collection Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
- 10. Overcoming Reading Challenges
  - Dealing with Digital Eye Strain
  - Minimizing Distractions
  - Managing Screen Time
- 11. Cultivating a Reading Routine Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Setting Reading Goals Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - o Carving Out Dedicated Reading Time

## **Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions**

- 12. Sourcing Reliable Information of Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Fact-Checking eBook Content of Electrolyte Data Collection Viscosity Of Nonaqueous Solutions I Alcohol Solutions
  - Distinguishing Credible Sources
- 13. Promoting Lifelong Learning
  - Utilizing eBooks for Skill Development
  - Exploring Educational eBooks
- 14. Embracing eBook Trends
  - Integration of Multimedia Elements
  - Interactive and Gamified eBooks

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