Self-consistent field theory of polymer-ionic molecule complexation

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A self-consistent field theory is developed for polymers that are capable of binding small ionic molecules (adsorbates). The polymer-ionic molecule association is described by Ising-like binding variables, $C_1^{\rm ion}(k\Delta)(=0 \text{ or } 1)$, whose average determines the number of adsorbed molecules, $n_{\rm HI}$ polymer gelation can occur through polymer-ionic molecule complexation in our model. For polymer-polymer cross-links through the ionic molecules, three types of solutions for $n_{\rm HI}$ are obtained, depending on the equilibrium constant of single-ion binding. Spinodal lines calculated from the mean-field free energy exhibit closed-loop regions where the homogeneous phase becomes unstable. This phase instability is driven by the excluded-volume interaction due to the single occupancy of ion-binding sites on the polymers. Moreover, sol-gel transitions are examined using a critical degree of conversion. A gel phase is induced when the concentration of adsorbates is increased. At a higher concentration of the adsorbates, however, a re-entrance from a gel phase into a sol phase arises from the correlation between unoccupied and occupied ion-binding sites. The theory is applied to a model system, poly(vinyl alcohol) and borate ion in aqueous solution with number of Physics. [doi:10.1063/1.3430745]

I. INTRODUCTION

The miscibility of neutral polymers and polyelectrolytes with ionic molecules in solvents has attracted much interest in polymer science over the last decades." In particular, phase separation induced by complexation between polymers and ionic molecules2-10 is widely studied in experiments11-14 and theory. 15-18 Noncovalent intermolecular interactions such as hydrogen bonding, dispersion, and electrostatic forces are often employed to bind smaller molecules onto polymers (adsorbents). For example, functionalized sidechains of polymers are capable of capturing relatively smaller molecules through these weak forces. 19,50 This association is typically sensitive to temperature, pH, and salt concentration. The reversibility of the complexation processes can be utilized for various potential applications in chemical engineering or supramolecular science to fabricate functional materials such as hydrogel.21

Adsorption phenomena are often described by the Langmuir model, where an adsorbate molecule (B) is adsorbed onto an unoccupied binding site (P) to form an adsorbed complex (BP). The Langmair model is based on the following assumptions: (1) the adjacent molecules adsorbed on the binding sites are mutually noninteracting. (2) the surface at which there are binding sites is geometrically uniform (i.e., not corrugated), and (3) each binding site is capable of adsorbing a single molecule. The number of adsorbed molecules is determined by the chemical equilibrium between nonadsorbed (free) adsorbates in bulk phase and adsorbed adsorbates. In this case, the association equilibrium is written as $B+P \rightleftharpoons BP$ (monocomplexation) with an equilibrium constant, K_{eq} . The Langmair equation is then given by

$$[BP] = \frac{[BP]_{max}K_{cq}[B]}{1 + K_{cq}[B]}.$$
 (1)

where $[BP]_{max}$ is the maximum value of [BP] as $[B] \rightarrow \infty$. It should be noticed that in general, an adsorbate molecule may be capable of binding to two (or more) binding sites. In this case, further complexation between BP and P can lead to interpolymer or intrapolymer association (dicomplexation) or gelation, $BP+P = BP_3$.

For polyelectrolytes, adsorption of monovalent and divalent counterions on charged sites occurs at low temperatures, effectively neutralizing the polyelectrolytes. In this case, a polyelectrolyte chain may collapse because the repulsive force between charged sites is weakened.^{23–26} Upon further addition of the divalent salts, the charges on the polyelectrolyte chain can be reversed as a result of an excess number of condensed counterions, corresponding to the charge inversion in colloidal science.²⁵

To characterize the Coulombic attractive force for adsorptions at a temperature, T, the Bjernim length, $I_B = e^2/4 \pi e k_B T$, and Coulomb coupling constant (or dimensionless inverse temperature), $\Gamma = I_B/\alpha$, are often employed. Here, e, e, k_B , and α denote the unit charge, dielectric constant, Boltzmann constant, and the distance between charged molecules, respectively. When $\Gamma > 1$, the complexation between charges occurs, indicating a strong coupling limit of the system. However, it is known that the Poisson–Boltzmann equation (PBE), a classical mean-field theory, fails to describe the

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Table of Contents Semi Empirical Self Consistent Field Molecular Theory Of Molecules

- 1. Understanding the eBook Semi Empirical Self Consistent Field Molecular Theory Of Molecules
 - The Rise of Digital Reading Semi Empirical Self Consistent Field Molecular Theory Of Molecules
 - Advantages of eBooks Over Traditional Books
- 2. Identifying Semi Empirical Self Consistent Field Molecular Theory Of Molecules
 - 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 Semi Empirical Self Consistent Field Molecular Theory Of Molecules
 - User-Friendly Interface
- 4. Exploring eBook Recommendations from Semi Empirical Self Consistent Field Molecular Theory Of Molecules
 - Personalized Recommendations
 - Semi Empirical Self Consistent Field Molecular Theory Of Molecules User Reviews and Ratings

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