

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, $C_i^{\text{ad}}(\Delta\Delta)(=0 \text{ or } 1)$, whose average determines the number of adsorbed molecules, n_{ad} . 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_{ad} 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 sodium chloride. Good agreement between theory and experiment is obtained. © 2010 American Institute 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.¹ In particular, phase separation induced by complexation between polymers and ionic molecules^{2–10} is widely studied in experiments^{11–14} 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 side-chains of polymers are capable of capturing relatively smaller molecules through these weak forces.^{19,20} 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 Langmuir 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 Langmuir equation is then given by

$$[BP] = \frac{[BP]_{\text{max}} K_{\text{eq}} [B]}{1 + K_{\text{eq}} [B]}, \quad (1)$$

where $[BP]_{\text{max}}$ 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, $BP + P \rightleftharpoons BP_2$.

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.^{22–24} 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 Bjerrum length, $l_B = e^2/4\pi\epsilon_0 k_B T$, and Coulomb-coupling constant (or dimensionless inverse temperature), $\Gamma = l_B/a$, are often employed. Here, e , ϵ , k_B , and a 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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Semi-empirical Self-consistent-field Molecular Orbital Theory of Molecules John Norman Murrell, Alan John Harget, 1972 *Semi-empirical Self-consistent-field Molecular Orbital Theory of Molecules* John Norman Murrell, A. J. Harget, 1972

Semiempirical Methods of Electronic Structure Calculation Gerald Segal, 2012-12-06 If one reflects upon the range of chemical problems accessible to the current quantum theoretical methods for calculations on the electronic structure of molecules one is immediately struck by the rather narrow limits imposed by economic and numerical feasibility. Most of the systems with which experimental photochemists actually work are beyond the grasp of ab initio methods due to the presence of a few reasonably large aromatic ring systems. Potential energy surfaces for all but the smallest molecules are extremely expensive to produce even over a restricted group of the possible degrees of freedom and molecules containing the higher elements of the periodic table remain virtually untouched due to the large numbers of electrons involved. Almost the entire class of molecules of real biological interest is simply out of the question. In general the theoretician is reduced to model systems of variable appositeness in most of these fields. The fundamental problem from a basic computational point of view is that large molecules require large numbers of basis functions whether Slater type orbitals or Gaussian functions suitably contracted to provide even a modestly accurate description of the molecular electronic environment. This leads to the necessity of dealing with very large matrices and numbers of integrals within the Hartree Fock approximation and quickly becomes both numerically difficult and uneconomic.

Energy Landscapes David Wales, 2003 The study of energy landscapes holds the key to resolving some of the most important contemporary problems in chemical physics. Many groups are now attempting to understand the properties of clusters, glasses and proteins in terms of the underlying potential energy surface. The aim of this book is to define and unify the field of energy landscapes in a reasonably self contained exposition. This is the first book to cover this active field. The book begins with an overview of each area in an attempt to make the subject matter accessible to workers in different disciplines. The basic theoretical groundwork for describing and exploring energy landscapes is then introduced followed by applications to clusters, biomolecules and glasses. In the final chapters, beautifully illustrated in full colour throughout, this book is aimed at graduate students and workers in the field.

Lecture Notes on Atomic and Molecular Physics ?akir Erko, Turgay Uzer, 1996 This book aims to present a unified account of the physics of atoms and molecules from a modern viewpoint. It is based on courses given by the authors at Middle East Technical University, Ankara and Georgia Institute of Technology, Atlanta, and is suitable for study at third and fourth year levels of an undergraduate course. Students should be able to read this volume and understand its contents without the need to supplement it by referring to more detailed discussions. The whole subject covered in this volume is expected to be finished in one semester.

Applying Molecular and Materials Modeling Phillip R. Westmoreland, Peter A. Kollman, Anne M. Chaka, Peter T. Cummings, Keiji Morokuma, Matthew Neurock, Ellen B. Stechel, Priya Vashishta, 2013-04-17

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Computational Medicinal Chemistry for Drug Discovery Patrick Bultinck, Hans De Winter, Wilfried Langenaeker, Jan P. Tollenare, 2003-12-17 Observing computational chemistry's proven value to the introduction of new medicines this reference offers the techniques most frequently utilized by industry and academia for ligand design. Featuring contributions from more than fifty pre eminent scientists *Computational Medicinal Chemistry for Drug Discovery* surveys molecular structure computation intermolecular behavior ligand receptor interaction and modeling responding to market demands in its selection and authoritative treatment of topics. The book examines molecular mechanics semi empirical methods wave function based quantum chemistry density functional theory 3 D structure generation and hybrid methods **Molecular Modeling for the Design of Novel Performance Chemicals and Materials** Beena Rai, 2012-03-23 Molecular modeling MM tools offer significant benefits in the design of industrial chemical plants and material processing operations. While the role of MM in biological fields is well established in most cases MM works as an accessory in novel products materials.

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and computational methods required to describe the experimentally observable behavior of the system By now many of the alternative theoretical approaches and computational procedures have been tested and intercompared More or less optimal methods for dealing with each aspect are emerging In many cases working equations even schematic algorithms have been developed with assumptions and caveats delineated **Scientific and Technical Aerospace Reports** ,1967

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