

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

Computational

molecular and materials modeling has emerged to deliver solid technological impacts in the chemical pharmaceutical and materials industries It is not the all predictive science fiction that discouraged early adopters in the 1980s Rather it is proving a valuable aid to designing and developing new products and processes People create not computers and these tools give them qualitative relations and quantitative properties that they need to make creative decisions With detailed analysis and examples from around the world Applying Molecular and Materials Modeling describes the science applications and infrastructures that have proven successful Computational quantum chemistry molecular simulations informatics desktop graphics and high performance computing all play important roles At the same time the best technology requires the right practitioners the right organizational structures and most of all a clearly understood blend of imagination and realism that propels technological advances This book is itself a powerful tool to help scientists engineers and managers understand and take advantage of these advances Quantum Nanochemistry, Volume One Mihai V. Putz,2016-03-30 Volume 1 of the 5

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Computational Modeling of Polymers Jozef Bicerano,1992-03-17 **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 development rather than a tool for direct innovation As a result MM engineers and practitioners are often seized with the question How do I leverage these tools to develop novel materials or chemicals in my industry Molecular Modeling for the Design of Novel Performance Chemicals and Materials answers this important question via a simple and practical approach to the MM paradigm Using case studies it highlights the importance and usability of MM tools and techniques in various industrial applications The book presents detailed case studies demonstrating diverse applications such as mineral processing pharmaceuticals ceramics energy storage electronic materials paints coatings agrochemicals and personal care The book is divided into themed chapters covering a diverse range of industrial case studies from pharmaceuticals to cement While not going too in depth into

fundamental aspects the book covers almost all paradigms of MM and references are provided for further learning The text includes more than 100 color illustrations of molecular models

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

Transition States of Biochemical Processes R. Gandour, 2013-06-29 The transition state theory has been from the point of its inception the most influential principle in the development of our knowledge of reaction mechanisms in solution It is natural that as the field of biochemical dynamics has achieved new levels of refinement its students have increasingly adopted the concepts and methods of transition state theory Indeed every dynamical problem of biochemistry finds its most elegant and economical statement in the terms of this theory Enzyme catalytic power for example derives from the interaction of enzyme and substrate structures in the transition state so that an understanding of this power must grow from a knowledge of these structures and interactions Similarly transition state interactions and the way in which they change as protein structure is altered constitute the pivotal feature upon which molecular evolution must turn The complete coupled dynamical system of the organism incorporating the transport of matter and energy as well as local chemical processes will eventually have to yield to a description of its component transition state structures and their energetic response characteristics even if the form of the description goes beyond present day transition state theory Finally the importance of biochemical effectors in medicine and agriculture carries the subject into the world of practical affairs in the use of transition state information for the construction of ultra potent biological agents

Electronic Structure Calculations on Graphics Processing Units Ross C. Walker, Andreas W. Goetz, 2016-04-18 Electronic Structure Calculations on Graphics Processing Units From Quantum Chemistry to Condensed Matter Physics provides an overview of computing on graphics processing units GPUs a brief introduction to GPU programming and the latest examples of code developments and applications for the most widely used electronic structure methods The book covers all commonly used basis sets including localized Gaussian and Slater type basis functions plane waves wavelets and real space grid based approaches The chapters expose details on the calculation of two electron integrals exchange correlation quadrature Fock matrix formation solution of the self consistent field equations calculation of nuclear gradients to obtain forces and methods to treat excited states within DFT Other chapters focus on semiempirical and correlated wave function methods including density fitted second order Møller Plesset perturbation theory and both iterative and perturbative single and multireference

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Rundle, 1956 **Computational Chemistry And Chemical Engineering - Proceedings Of The Third Unam-cray Supercomputing Conference** Gerardo Cisneros, Juan Antonio Cogordan, Miguel Castro, Chumin Wang, 1997-10-31 This book provides a wide ranging and up to date description of state of the art computational methodologies in chemistry and chemical engineering It displays a representative mix of topics on the computation and modeling of chemical systems of all sizes from the very small atomic to the very large industrial The book constitutes an excellent overview for graduate students as well as a critical update for researchers **Carbon Bonding and Structures** Mihai V. Putz, 2011-08-27 Carbon Bonding and

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