

Self-consistent field theory of polymer-ionic molecule complexation

Issei Nakamura^{ab} and An-Chang Shi^{2a}*Department of Physics and Astronomy, McMaster University, 1280 Main Street West, Hamilton, Ontario L8S 4L8, Canada*

(Received 14 March 2010; accepted 27 April 2010; published online 17 May 2010)

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, $l_B = e^2/4\pi\epsilon_0\epsilon k_B T$, 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

^aElectronic mail: nakamura@caltech.edu. Present address: Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, California 91125, USA.

^bElectronic mail: shi@mcmaster.ca.

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