

NANOSCALE STRUCTURE, PHASE BEHAVIOR AND MACROSCOPIC PROPERTIES OF BIOPOLYMER GELS

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Soft condensed matter such as polymers, colloids and other complex fluids deliver unique functionality and utility for applications ranging from the nanoscopic (e.g., nanotubes and nanofibers) to the microscopic (e.g., drug delivery microcapsules) and macroscopic (e.g., medical implants and high performance engineering composites) length scales. Biological systems operate at the cellular and subcellular levels; therefore, material properties including structure, thermodynamic and mechanical properties must be determined to dimensions below 100 nm. Synthetic polymers have been recently designed to mimic the hierarchical structure and function of biological macromolecules, such as DNA, proteins, as well as biological membranes and cells. Understanding the underlying physical characteristics of these systems enables molecular and nanometer scale manipulation with the aim of engineering useful and novel properties.

One main focus of our research is on the roles that nanoscale structures and interactions play in determining the macroscopic properties of polyelectrolyte gels. In the physiological environment biopolymers are exposed to different ions. Structure formation is the result of a complex interplay between thermodynamic and kinetic factors that depends on the interactions between ions and polymer molecules. In many polyelectrolyte gels a discontinuous volume transition is observed upon the introduction of multivalent ions. The complexity of the behavior of charged macromolecular systems makes necessary to investigate the structure and physical properties on all length scales from the atomic level up to the macroscopic level. We developed a multiscale approach to examine structural hierarchy, phase behavior and macroscopic properties in biopolymer solutions and gels.

To illustrate our approach, experimental results will be presented that provide a critical missing link between the thermodynamic properties and the nanoscale structure from which these properties ultimately originate. Scattering measurements (SAXS, SANS, and light scattering) are made that probe structures over a wide range of length scales (1-500 nm) and provide insight into the hierarchical organization of the molecules and the phase behavior of the polymer system. Osmotic measurements are used to relate the macroscopic properties to the morphology. We complement the experimental findings with molecular dynamics simulations made on model polyelectrolyte networks containing mono- and multivalent counterions. The roles of the virial contributions to the osmotic pressure are determined. The interplay between the electrostatic energy and the counterion excluded-volume entropy appears to be responsible for the discontinuous volume transition observed in a variety of polyelectrolyte gels.

These studies provide the insight essential to design and control gel properties by tailoring the chemical and nanometer scale structures and interactions.

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