

Stony Brook University The Graduate School

Doctoral Defense Announcement

Abstract

Rheology and Structure of Thermoreversible Hydrogels

By

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Highly concentrated solutions of non-ionic amphiphilic triblock copolymer poly(ethylene oxide)₉₉-poly(propylene oxide)₆₇-poly(ethylene oxide)₉₉ (Pluronic F127) are widely used in numerous biomedical applications, such as drug delivery vehicles, gels for replacing biological fluids such as synovial fluid and nucleus pulposus, and surfactants for emulsification of food and personal care products. The Pluronic copolymers are popular for these applications, since their gelation properties are thermoreversible and easily controlled by varying the concentration. They are liquid below room temperature and gel at body temperature.

In this dissertation, thermal gelation and structure of high concentration triblock copolymer Pluronic F127-clay aqueous solutions were characterized by rheological measurements, differential scanning calorimetry (DSC) and small angle X-ray/neutron scattering. Small angle neutron scattering (SANS), under shear using a Couette cell in radial and tangential scattering geometry, was performed to examine the structural evolution of the polymeric micellar macro-lattice formed by concentrated aqueous solutions of triblock copolymer-Pluronic F127, as a function of the shear rate. The micellar gel showed a shear thinning by forming a layered stacking of two-dimensional hexagonally close packed (HCP) polymer micelles. A theoretical model was developed to calculate 2D SANS scattering patterns that can be compared with the experimental data.

In order to improve the mechanical properties of the gel, while still maintaining the thermo-reversibility, we synthesized multiblock structures, where the F127 construct would be repeated several times. The rheological and structural properties of the gels were characterized as a function of temperature, composition and degree of polymerization. For F127 solutions just below their gel point, substitution of F127 with as little as 1% multiblock succeeded in forming a physical gel. Percolation theory was used to understand the modulus growth when multiblock was added to F127 solutions, assuming the multiblocks form bridges between adjacent micelles.

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