81b Stimuli-Responsive Lysine-Glycine Block Copolypeptide Supramolecular Structures

Daniel F. Shantz and Jeng-Shiung Jan

Studies of the self-assembly of poly(L-lysine)-b-polyglycine diblock and triblock copolypeptides in aqueous solution will be presented. Dynamic light scattering (DLS), confocal microscopy, and circular dichroism (CD) were used to characterize the size, supramolecular structures, and folded states of the block copolypeptides at different solution conditions. The block copolypeptides form true equilibrium structures that can be manipulated between vesicles and micelles based on solution conditions, and the stimuli to induce these transformations can be pH, salt, or anions (for example, perchlorate anion). These structural interconversions are shown to be primarily due to the change in conformation of the lysine block and are completely reversible. Preliminary work is also reported demonstrating that the non-covalent supramolecular structures formed can be converted to covalent supramolecular structures and their swelling behavior regulated by changes in pH or ionic strength, and that these block copolypeptides can be used to mineralize silica at ambient conditions. The results of this work show that these previously unreported materials have unique properties of relevance to numerous applications including drug delivery, controlled released, encapsulation, and biomineralization/biomimetic syntheses of hard matter.

Results will also be presented describing the self-assembly behavior of lysine-alanine and lysine-(glycine,alanine) block copolypeptides. The latter represent the ability to incrementally increase polypeptide hydrophobicity as compared to the lysine-glycine polypeptides. The former materials represent a bridge to the work by Deming and coworkers on lysine-leucine and lysine-valine block copolypeptides that have been shown to form hydrogels. Preliminary results on lysine-alanine block copolypeptides indicate that much higher polypeptide concentrations are needed to form hydrogels (~10 wt% for K100A25) as compared to the corresponding lysine-leucine copolymers (< 1 wt%).