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Solution Self-Assembly of Globular Protein-Polymer Conjugate Block Copolymers[1]

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Controlling the nanostructured self-assembly of globular proteins and enzymes can significantly advance the applications of soft materials as catalysts, sensors, and medical materials. However, the incorporation of globular proteins as one block in the block copolymer introduces changes in chain shape, chain entropy, and specific interactions that significantly impact the thermodynamics of self-assembly. Here, we explore the self-assembly of model globular protein-polymer block copolymers in concentrated solutions to form nanostructured materials. A phase diagram as a function of concentration and temperature for a model material mCherry-poly(N-isopropylacrylamide) (PNIPAM) is asymmetric, showing hexagonal cylinders for coil fractions less than 0.5 and a lamellar ordering for coil fractions greater than 0.5, divided by a narrow region of hexagonally perforated lamellae. Order-order transitions as a function of temperature are driven by the thermoresponsive desolvation of PNIPAM. Surprisingly, the materials exhibit reentrant order-disorder transition behavior, such that the conjugate block copolymers are disordered at both low and high concentrations but well-ordered at intermediate concentrations. Changing the polymer chemistry to monomers with different types of hydrogen bonding results in significant changes in the self-assembly, including the observation of a cubic phase that shows the same scattering pattern as the gyroid phase observed in traditional block copolymers. The choice of polymer also has a strong impact on the order-disorder transition concentration, demonstrating that the polymer-protein interaction plays a significant role in governing self-assembly in solution. Consistent with this effect, the order-disorder transition concentration is minimized in symmetric conjugates. Changing the protein from mCherry to myoglobin results in a reduction in ordering, suggesting that the regularity of the protein shape is important.

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