Molecular Engineering of Polyelectrolyte Complex Materials

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Understanding how chemical sequence affects the self-assembly of polymers is a grand challenge across materials science and biology. The complexity of a near infinite number of sequence permutations is made even more difficult because of the convergence of interactions across a wide range of length-scales. In particular, electrostatic interactions are long-range, and compete with shorter-range effects such as van der Waals forces, hydrogen bonding, etc. We draw inspiration from membraneless organelles, which involve sequence-specific intrinsically disordered proteins that interact via weak, multivalent interactions to achieve transient liquid-liquid phase separation in cells. The resulting 'membraneless organelles' have also been shown to enable the selective uptake of specific enzymes. We utilize polypeptides as model sequence-controlled polymers to study how the patterning or presentation of charges and other chemical functionalities can modulate the potential for liquid-liquid phase separation via complex coacervation. We further examine how the distribution of charge on globular proteins can be used to facilitate selective uptake into coacervate phases, and how such materials can be used to stabilize proteins and viruses against denaturation. This molecular-level understanding of polyelectrolyte complexation is further enhanced by detailed thermodynamic and rheological characterization. Our experimental efforts are supported by the parallel development of computational approaches for modeling and predicting the phase behavior of patterned polymeric materials. Our goal is to establish molecular-level design rules to facilitate the tailored creation of materials based on polyelectrolyte complexation that can both illuminate self-assembly phenomena found in nature, and find utility across a wide range of real-world applications.