Mesoscale materials design for bioinspired architectures

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A hallmark of Natural materials with remarkable properties is hierarchical design, building from the molecular scale through precise mesoscale architectures to full, multifunctional composites at the macroscale. For example, tendon assembly spans amino acids to mesoscale fibrils to the macrostructure, a high-modulus hydrogel that readily adheres to bone to bear and transmit load. Similarly, ecribellate capture silk follows a hierarchy of amino acids to proteins to mesoscale fibers, capturing prey by spooling inside droplets when compressed and exhibiting solid-like elasticity in tension. Despite such compelling inspiration, few synthetic mesoscale materials exhibit similar sophistication due to the combined challenges of embedding coexisting functionalities and fabricating precise structures. This presentation will describe i) the design and preparation of a class of structures that are built from the scale of individual monomers to multifunctional copolymers to mesoscale ribbons, then ii) their use in replicating several architectures found in Nature. Key examples include capillary-driven wrapping of mesoscale polymer ribbons around oil-in-water droplets and flagellum-like head-tail assemblies featuring polymer ribbons as “arms” extended from the droplet surface. To better understand the observed ribbon-droplet adhesion behavior, parameters including the peel force *Fc* and work of adhesion *G* were quantified as a function of ribbon compliance and composition by deflection of a carbon fiber cantilever. Together, the presented structures and quantitative analysis afford a simple materials platform that underlies a new class of useful and bio-inspired materials.