The ability to create synthetic materials that mimic the structural and mechanical properties of native tissues is crucial for scaffolding applications. Unfortunately, design of such materials remains a significant challenge. Many native tissues and synthetic soft biomaterials are comprised of polymer networks. What are the “minimum physics” that we must build into a synthetic polymer network in order to reproduce the array of mechanical and structural properties exhibited by biological gels? I will discuss this issue, first focusing on engineering crystalline domains into associative hydrogels of poly(lactic acid)-poly(ethylene oxide)-poly(lactic acid) (PLA-PEO-PLA) triblock copolymers. In aqueous media, these materials form associative gels with PLA domains serving as network junctions. We can create nanoscale crystalline junctions via copolymers in which the PLA block is poly(L-lactide) (PLLA), or amorphous junctions through copolymers in which the PLA blocks contain a racemic mixture of D-lactide and L-lactide (PRLA). These crystalline junction points yield biocompatible gels with elastic moduli that are an order of magnitude higher than previously reported with similar associative gels. Additionally, these domains appear to be responsible for self-assembly of interesting nano- to micro-scale structural features in the gels, similar to those appearing in block polypeptide networks. Finally, we have demonstrated that the PLLA triblocks exhibit a novel morphology, disklike micelles. This morphology had previously only been theorized to be accessible with ionic copolymers and had only been experimentally observed with ABC triblocks in the “superstrong segregation” regime. In the latter half of the talk, I will discuss recent results on PEG hydrogels with a nearly ideal tetrafunctional structure. We believe the low level of defects in this network structure lead to hydrogels with an extremely high resilience, reminiscent of elastin and resilin. Collectively, our work shows that tacticity and topology can be used to access novel structures and properties in relatively simple synthetic polymers, perhaps giving insight into some of the routes Nature uses to create complex soft materials.

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