Organic Division Seminar

Organocatalysis: Strategies for Controlled Polymerization

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We have developed a family of organocatalysts for the controlled polymerization of lactones to well-defined polyesters of novel architectures and macromolecular topologies. These organocatalysts are both selective and highly active, exhibiting characteristics of living polymerization with turnover frequencies as high as 18 per second with turnover numbers up to 1000. In the presence of alcohols, N-heterocyclic carbenes are potent catalysts for the ring-opening polymerization of lactide to generate linear polylactides. In the absence of alcohols, N-heterocyclic carbenes mediate the zwitterionic polymerization of lactide or caprolactone to generate high molecular weight cyclic polyesters of narrow molecular weight distribution. The zwitterionic polymerization of lactones provides a new strategy for generating high molecular weight cyclic polyesters of defined molecular weight and molecular weight distribution. These cyclic polyesters exhibit different properties than their linear homologs by virtue of the topological constraint of connected chain-ends. Polymerization of caprolactone generates crystalline cyclic polyesters, enabling investigations on the role of the cyclic topology on the crystalline structure and morphology. The combination of organocatalytic ring-opening polymerization with living free radical methods generates a variety of novel architectures, including linear and branched amphiphilic block copolymers. Metal-free block polymers generated with organic catalysts assemble into nanostructures that encapsulate drugs and biological probes. The unusual stereocomplexation phenomena characteristic of enantiomeric polylactides directs their assembly of block copolymers; we have generated a new family of thermoresponsive mixed micelles that contain two different polymeric surfactants. The exceptional control of these catalytic systems provides the opportunity to design the structure, function and assembly of the resultant biodegradable polymers and assemblies.