

Molecular Level Design of Mechanically Interlocked and Supramolecular Polymers for Energy Storage Applications

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The alarming environmental issues originating from the continuous consumption of fossil fuels have triggered the large-scale utilization of renewable energy sources, which should be paired with high energy density storage systems due to their intermittent nature. Unfortunately, the limited energy densities of the current storage technologies such as lithium-ion batteries (LIBs) and supercapacitors are slowing down the transition to green sustainable energy. Thus, switching to advanced electrode material chemistries that possess higher theoretical capacity will increase the storage capacity and practical energy density such as silicon (Si), lithium metal (Li) anodes, and sulfur cathodes. However, these high energy density electrodes suffer from severe drawbacks hindering their applications. In this direction, the thesis focused on the fundamental understanding of failure mechanisms of these electrodes along with the molecular-level design of polymeric materials to address these issues.

Chapter 2&3: We utilized mechanically interlocked polymers in the form of topological crosslinked polyrotaxane to stabilize the volume variations of Si anode composite and endure the topological deformation of Li metal host; thus, leading to excellent cycling stability.

Chapter 4: We developed new charged porous conducting covalent triazine-based frameworks (CTFs) as hosts for sulfur and to immobilize lithium polysulfides shuttling through ion-dipole interactions within the pores and enable high mass loadings in lithium-sulfur batteries.

Chapter 5: We discovered a new dual-functional high donor electrolyte to enable operation under lean electrolyte conditions and high mass loadings lithium-sulfur batteries.

Chapter 6: In an effort to understand the factors affecting each storage mechanism to improve the electrochemical capacitance, we designed tailor-made porous CTFs with a precise control over each factor to reveal their contribution to maximize specific capacity.

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