

Functional Protective Coatings Based on Polysaccharides and Single-ion Conducting Polymers for Metal Batteries

The practical application of lithium-metal batteries (LMBs) is limited by the intrinsic instability of Li metal, which leads to uncontrolled lithium growth, electrolyte decomposition, and mechanically fragile interfaces. This thesis addresses these challenges by developing polysaccharide and single-ion-conducting polymer (SICP)-based coatings that stabilize the Li-electrolyte interface, regulate Li⁺ transport, and promote uniform plating. These coatings improve interfacial stability, lithium utilization, and cycling reversibility in both liquid and solid-state electrolytes. The thesis is divided into three sections. First, synthetic pathways for polysaccharide-based materials are explored. Cellulose derivatives exhibited good mechanical properties but limited reactivity and solubility, while cyclodextrins enabled the successful incorporation of lithium-conducting groups. The second section focuses on coating formulation and mechanism study. A multifunctional blend of TDMSC, P(LiMTFSI), and LiNO₃ forms a conformal, ionically conductive layer that suppresses high-surface-area lithium formation. Combined electrochemical, surface, and interphase analyses reveal a porous, electrolyte-permeable interface that regulates Li⁺ flux and limits electrolyte decomposition. Finally, the coating strategy is extended to magnesium metal. TDMSC-P(LiMTFSI) films improve magnesium morphology and utilization, demonstrating partial transferability while highlighting challenges associated with divalent ions.