Abstract

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The ongoing global transition toward sustainable energy has intensified research efforts into high-performance and environmentally benign energy storage systems. Lithium-ion batteries (LIBs), with their superior energy density and long cycling life, remain the dominant technology for a wide range of applications, including consumer electronics, electric vehicles (EVs), and grid-scale storage. However, the widespread use of environmentally harmful, non-water-soluble electrode binders, such as poly(vinylidene fluoride) (PVDF), raises significant concerns due to their toxicity, high processing costs, and limited recyclability. In response, the development of water-soluble, biodegradable binder materials has become a critical focus for high electrochemical performance of next-generation LIBs.

This research begins by establishing the foundational background of LIBs, emphasizing the crucial role of binders in ensuring electrode cohesion, structural integrity, and stable electrochemical behavior during long cycling. Traditional binders are reviewed in detail, followed by an in-depth discussion of recent advancements. The limitations of current binder materials motivate the development of pectate-based PIL binders proposed in this thesis.

1-allyl-3-methylimidazolium pectate ([AMIm][Pectate]) is synthesized by neutralizing polygalacturonic acid (pectic acid) with [AMIm][OH]. The obtained binder exhibits strong interfacial adhesion due to the abundant hydroxyl (-OH) groups on the pectate polymer backbone, which interact favorably with both graphite and conductive additives. Meanwhile, the imidazolium cation enhances Li^+ mobility, contributing to improved charge-transfer kinetics. Electrochemical testing of graphite anodes assembled with [AMIm][Pectate] demonstrates enhanced cycling stability and capacity retention compared to PVDF-based controls. Scanning electron microscopy (SEM) and X-Ray photoelectron spectroscopy (XPS) further confirm the binder's ability to maintain electrode integrity over long cycling.

The synthesis of [Choline][Pectate] is via the neutralization of pectic acid with choline hydroxide ([Choline][OH]). This binder is evaluated under even more demanding conditions, including high current densities (1 C, 2 C, 5 C) and full-cell configurations pairing graphite anodes with $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ (NCM) cathode. Compared to CMC-SBR binder, [Choline][Pectate] exhibits superior rate capability and coulombic efficiency, attributed to its enhanced ionic conductivity and mechanical robustness. Post-mortem analyses of cycled electrodes reveal minimal delamination and stable solid electrolyte interphase (SEI) formation, further validating its practical viability.

Finally, these findings not only demonstrate the electrochemical viability of [AMIm][Pectate] and [Choline][Pectate] as sustainable alternatives to non-water-soluble binders but also establish a versatile platform for ongoing binder innovation. The integration of pectate-derived polymer backbones with tunable ionic liquid components offers a promising pathway for the development of next-generation binder systems. Moreover, this work makes a significant contribution to the field of LIB materials by introducing a green, scalable, and high-performance binder strategy aligned with global sustainability objectives.

Keywords: Lithium-ion battery, Graphite anode, Binder, Water soluble, High electrochemical performance.