Recent Advancements in Solid Electrolytes for Rechargeable Solid-State Batteries

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Extended Abstract

The fundamentally safe nature and the potential of higher energy density of solid-state batteries (SSBs) than Li-ion batteries (LIBs) have spurred enormous research activities on SSBs worldwide for the last 20 years. A key component for SSBs is a solid electrolyte that can meet multi-functional requirements, including high ionic conductivity, wide electrochemical window, being stable with both cathodes and anodes, good film-forming ability and high flexibility for conformal physical contact with electrodes, and easy processing with low cost. In this presentation, we will highlight the research efforts to pursue solid electrolytes in the NSF Center of All-Solid-State Batteries at Illinois Tech. We have investigated synthesis and properties of amorphous/crystalline Li₃OCl electrolyte, 1.6Li₂O-TaCl₅ oxychloride electrolyte, and polyethylene oxide (PEO) electrolyte with covalent organic framework (PEO/COF). We find that hydrothermal synthesis with rapid removal of OH⁻ and H⁺ ions as steam from water-solvated Li₃OCl can result in amorphous Li₃OCl electrolyte, while amorphous 1.6Li₂O-TaCl₅ oxychloride electrolyte can be formed via high-energy ball milling at room temperature (RT). Amorphous Li₃OCl and 1.6Li₂O-TaCl₅ oxychloride as well as PEO/COF electrolytes all possess high ionic conductivities in the order of 10^{-3} S/cm at RT. Furthermore, amorphous Li₃OCl is stable against Li⁺/Li electrode and has oxidation resistance up to 5 V vs. Li⁺/Li, while amorphous 1.6Li₂O-TaCl₅ oxychloride is not stable against Li⁺/Li electrode, but stable above 5 V vs. Li⁺/Li. PEO/COF electrolytes also possess oxidation resistance up to 5 V vs. Li⁺/Li. SSBs assembled using these electrolytes have exhibited good cycle stability at RT, demonstrating the potentials of these novel solid electrolytes in the development of SSBs in the near future.