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Anodic Stability of Li_6PS_5Cl/PEO Composite Polymer Electrolytes for All-Solid-State Lithium Batteries: A First-Principles Molecular Dynamics Study

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Abstract : All-solid-state lithium batteries (ASSLBs) are increasingly recognized as a safer and more reliable alternative to conventional lithium-ion batteries due to their non-flammable nature and enhanced safety performance. ASSLBs utilize a range of solid-state electrolytes, including solid polymer electrolytes (SPEs), inorganic solid electrolytes (ISEs), and composite polymer electrolytes (CPEs). SPEs are particularly valued for their flexibility, ease of processing, and excellent interfacial compatibility with electrodes, though their ionic conductivity remains a significant limitation. ISEs, on the other hand, provide high ionic conductivity, broad electrochemical windows, and strong mechanical properties but often face poor interfacial contact with electrodes, impeding performance. CPEs, which merge the strengths of SPEs and ISEs, represent a compelling solution for next-generation ASSLBs by addressing both electrochemical and mechanical challenges. Despite their potential, the mechanisms governing lithium-ion transport within these systems remain insufficiently understood. In this study, we designed CPEs based on argyrodite-type Li_6PS_5Cl (LPSC) combined with two distinct polymer matrices: poly(ethylene oxide) (PEO) with 24.5 wt% lithium bis(trifluoromethane)sulfonimide (LiTFSI) and polycaprolactone (PCL) with 25.7 wt% LiTFSI. Through density functional theory (DFT) calculations, we investigated the interfacial chemistry of these materials, revealing critical insights into their stability and interactions. Additionally, ab initio molecular dynamics (AIMD) simulations of lithium electrodes interfaced with LPSC layers containing polymers and LiTFSI demonstrated that the polymer matrix significantly mitigates LPSC decomposition, compared to systems with only a lithium electrode and LPSC layers. These findings underscore the pivotal role of CPEs in improving the performance and longevity of ASSLBs, offering a promising path forward for next-generation energy storage technologies.

Keywords : all-solid-state lithium-ion batteries, composite solid electrolytes, DFT calculations, Li-ion transport
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