

Characterization of Ion Transport near the Electrolyte/Cathode Interface within Lithium-Ion Battery: A Molecular Dynamics Simulation Study

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To improve the performance of lithium-ion battery (LIB), various studies have been developing functional binders to reduce the electrolyte/electrode interfacial impedance, R_{int} . Here, we utilized molecular dynamics (MD) simulations to characterize Li^+ transport at the liquid electrolyte/ LiFePO_4 (LFP) cathode interface and the molecular effects of functional binders. Compared with conventional poly(vinylidene fluoride) (PVDF), Li^+ -coordinating or negatively charged polymers can lower the Li^+ free energy at interface and effectively reduce R_{int} . Yet, the reduction of R_{int} may be limited by the polymer chain rigidity. From non-equilibrium MD, we identified three sub-interfacial regions during the Li^+ (de-)intercalation: conventional Stern and diffusive layers, and a unique Li^+ depletion region. Analyses suggest that the concentration gradient and the electric field drive the Li^+ transport in the Stern and diffusive layers but not in the depletion region. The combined results suggest that improving the Li^+ affinity and the local mobility at interface can reduce R_{int} , where the free energy and electric potential variations play dominant roles.