

The Synergistic Effect of Cation and Anion of an Ionic Liquid Additive for Lithium Metal Anodes

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Lithium metal anodes are steadily gaining more attention, as their superior specific capacities and low redox voltage can significantly increase the energy density of rechargeable batteries far beyond those of current Li-ion batteries (LIBs). Nonetheless, the relevant technology is still in a premature research stage mainly due to the uncontrolled growth of Li dendrites that ceaselessly cause unwanted side reactions with electrolyte.

In order to circumvent these drawbacks, it is essential to suppress the dendrite growth and keep the solid-electrolyte-interphase (SEI) layer stable. Toward these ends, attempts have been made, such as incorporating artificial SEI layers,[1] polymer electrolytes,[2] solid electrolytes,[3] and porous nanostructures.[4] However, the majority of these approaches either require further improvements in electrochemical performance or are not scalable.

Here, we report an IL additive consisting of 1-dodecyl-1-methylpyrrolidinium (Pyr1(12)⁺) cation and bis(fluorosulfonyl)imide (FSI⁻) anion, in an attempt to heighten the technological maturity of Li metal anodes. The Pyr1(12)⁺ cation engages an electrostatic shielding effect to distribute the concentration of Li ion flux uniformly over the electrode, whereas the FSI⁻ anion can induce the formation of rigid SEI layers.[5] Furthermore, the nonpolar aliphatic chain attached to the pyrrolidinium adds a 'lithiophobic' effect, which further interferes with the Li ion adsorption to the dendrites. The ultimate effect of the IL is based on the synergy of the cation and anion, as they independently improve chemical and physical properties. A series of control ILs with modified cation and anion structures elucidate the respective roles of the cation and anion of the current IL of interest. From a practical viewpoint, the present IL serves as an electrolyte additive such that a relatively small amount (molar ratio between IL and solvent=1/12) was added to a conventional electrolyte solution, 1 M lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) in dioxolane/dimethoxyethane (DOL/DME), and the inherent shortcomings of ILs are thus effectively avoided.

References:

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