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 batteries far beyond those of current Li-ion batteries. 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 this shortcoming, herein, we report an ionic liquid (IL) additive, namely 1-dodecyl-1-methylpyrrolidinium ($Pyr1(12)^+$) bis(fluorosulfonyl)imide (FSI^-), for conventional electrolyte solutions. The $Pyr1(12)^+$ cation with a long aliphatic chain mitigates dendrite growth via the combined effects of electrostatic shielding and lithiophobicity, whereas the FSI^- anion can induce the formation of rigid SEI layers. The synergy between the cation and anion significantly improves cycling performance in asymmetric and symmetric control cells and a full cell paired with a $LiFePO_4$ cathode. The present study provides a useful insight into the molecular engineering of electrolyte components by manipulating the charge and structures of the involved molecules.