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Stable Conversion Chemistry-Based Lithium Metal Batteries Enabled by Hierarchical Multifunctional Polymer Electrolytes with Near-Single Ion Conduction

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Abstract

The low Coulombic efficiency and serious safety issues resulting from uncontrollable dendrite growth have severely impeded the practical applications of lithium (Li) metal anodes. Herein we report a stable quasi-solid-state Li metal battery by employing a hierarchical multifunctional polymer electrolyte (HMPE). This hybrid electrolyte was fabricated via in situ copolymerizing lithium 1-[3-(methacryloyloxy)propylsulfonyl]-1-(trifluoromethanesulfonyl)imide (LiMTFSI) and pentaerythritol tetraacrylate (PETEA) monomers in traditional liquid electrolyte, which is absorbed in a poly(3,3-dimethylacrylic acid lithium) (PDAALi)-coated glass fiber membrane. The well-designed HMPE simultaneously exhibits high ionic conductivity (2.24 × 10-3 S cm−1 at 25 °C), near-single ion conducting behavior (Li ion transference number of 0.75), good mechanical strength and remarkable suppression for Li dendrite growth. More intriguingly, the cation permselective HMPE efficiently prevents the migration of negatively charged iodine (I) species, which provides the as-developed Li-I batteries with high capacity and long cycling stability.