Nano-confinement engineered glassy MOF electrolytes for high-performance lithium metal batteries

Phd Jiaqi Huang[1], Dr. Yin Hu[1], Prof. Ulla Lassi[2], Prof. Xiaoyan Ji[1]
[1]Luleå University Of Technology, , Sweden, [2]University of Oulu, , Finland

Composite-solid-state electrolytes (CSEs) hold significant promise for the practical application of lithium metal batteries (LMBs). However, insufficient interfacial coupling between polymer matrices and inorganic fillers results in heterogeneous ion-transport pathways, space-charge, and mechanical discontinuity, thereby limiting Li+ mobility and electrochemical stability. Herein, we report a nanoscale-confined polymer electrolyte constructed within a glassy metal-organic framework, establishing an isotropic, grain-boundary-free ion transport network with enhanced mechanical robustness and intrinsic nonflammability. The resulting electrolyte exhibits rapid Li+ transport, delivering a high ionic conductivity of 1.2 × 10−3 S cm−1, and an elevated lithium-ion transference number (tLi+) of 0.74, together with excellent tensile strength and elongation (1.2 MPa and 167%, respectively). Symmetric Li||Li cells demonstrate stable Li plating/stripping for up to 1600 h at 0.2 mA cm−2, while Li||LiFePO4 cells maintain long-term cycling stability over 900 cycles at room temperature. Benefiting from a wide electrochemical stability window of up to 5.1 V, Li||LiNi0.8Co0.1Mn0.1O2 cells deliver stable cycling over 240 cycles at 2 C under 4.2 V and maintain stable operation for 100 cycles at an elevated cut-off voltage of 4.4 V, highlighting the robustness of the electrolyte under high-voltage conditions.