Dynamic physical network constructed by tripartite H-bonds in artificial SEI to shape dendrite-free lithium-metal anode

Qingping Wu[1,2], Yuhan Mei[2], Houyang Chen[2] and Yan Lu[1,3,4]

[1] Institute of Electrochemical Energy Storage, Helmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner-Platz 1, 14109 Berlin, Germany, [2] Chongqing Institute of Green and Intelligent Technology, Chinese Academy of Sciences, Chongqing 400714, China, [3] Institute for Technical and Environmental Chemistry, Friedrich-Schiller-Universität Jena, Philosophenweg 7b, 07743 Jena, Germany, [4] Helmholtz Institute for Polymers in Energy Applications (HIPOLE Jena), Lessingstraße 12-14, 07743 Jena, Germany

Stabilizing the composition of solid electrolyte interfaces (SEIs) and strengthening its structure are necessary conditions to ensure the safety and long lifespan of Lithium-Metal Batteries (LMBs)[1-2]. Herein, we designed dynamic physical network (DPN) with abundant lithiophilic/anionphilic groups through tripartite hydrogen bonds (H-bonds), serving as artificial SEIs in high-loading NCM811-Li metal batteries.[3] The formation and dissociation of DPN endowed by tripartite H-bonds under tension release during Li plating/stripping cycling skillfully balance the contradiction between mechanical robustness and deformability. Li-O bonds between lithiophilic sites and Li ions, and extra H-bonds between hydroxyl and electrolyte anions, endow DPN with functions of homogenizing Li-ion flux and accelerating its desolvation, synergistically achieving the uniform Li-ion deposition and weakening the charge shielding. The artificial SEI enhanced Li-anode (DPN@Li) withstood repeated Li ions plating/stripping processes for over 1000 h, which is 5 times longer than pure Li-anodes, and maintained low overpotential at a high current density of 10 mA cm^{-2}. DPN@Li-based NCM811 full cells deliver high specific capacities and outstanding cycle life over 3000 cycles. Further, DPN@Li possesses excellent electrochemical performance in the high active material loading (7.84 mg cm^{-2}) and foldable pouch cells. This work provides a conceptual framework of DPN constructed by multiple weak intermolecular interaction for artificial SEI to shape anode performance, and achieves the idea with a facile manner and simple chemical substances to promote practical applications of LMBs.

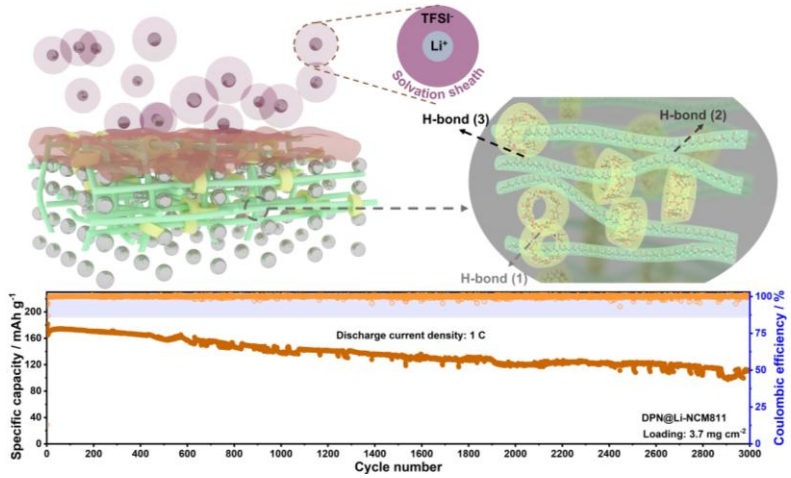

Figure. Schematic illustration of DPN in artificial SEI to shape dendrite-free LMBs

References:

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