Orientated Deposition of Li_2S for Fast-Charging Lithium-Sulfur Batteries

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Precipitation/dissolution of insulating Li_2S has long been recognized as the rate-determining step in lithium–sulfur (Li–S) batteries, which dramatically undermines sulfur utilization at elevated charging rates. Herein, we present an orientated Li_2S deposition strategy to achieve extreme fast charging (XFC, ≤15 min) through synergistic control of porosity, electronic conductivity, and anchoring sites of electrode substrate [1]. Via magnesiothermic reduction of a zeolitic imidazolate framework, a nitrogen-doped and hierarchical porous carbon with highly graphitic phase was developed. This design effectively reduces interfacial resistance and ensures efficient sequestration of polysulfides during deposition, leading to (110)-preferred growth of Li_2S nanocrystalline between (002)-dominated graphitic layers. Our approach directs an alternative Li_2S deposition pathway to the commonly reported lateral growth and 3D thickening growth mode, ameliorating the electrode passivation. Therefore, a Li–S cell capable of charging/discharging at 5C (12 min) while maintaining excellent cycling stability (82% capacity retention) for 1000 cycles is demonstrated. Even under high S loading (8.3 mg cm^{-2}) and low electrolyte/sulfur ratio (3.8 μL mg^{-1}), the sulfur cathode still delivers a high areal capacity of >7 mAh cm^{-2} for 80 cycles.

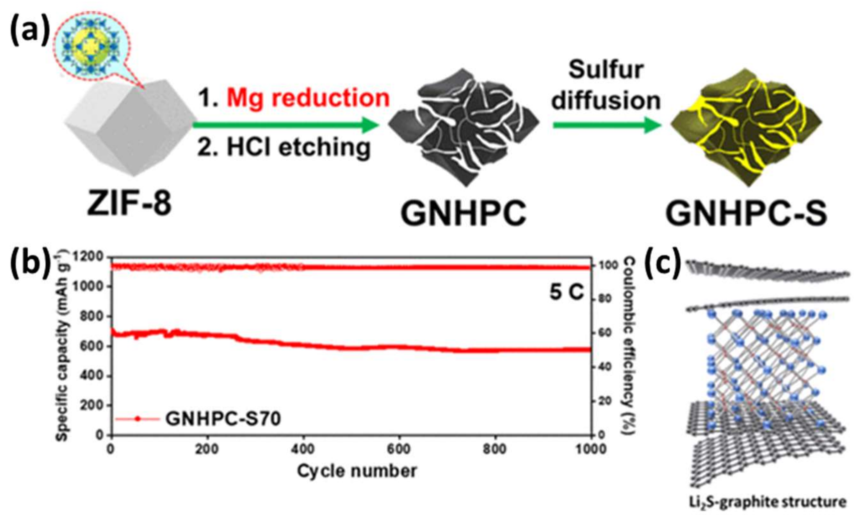

Figure 1. (a) A schematic illustration of synthetic procedures for carbon host and sulfur loading. (b) Long-term cycling performance of the GNHPC-S70 at a high current density of 5 C for 1000 cycles. (c) A schematic illustration showing the expansion of graphite layer and intergrowth structure, facilitating both ionic/electronic conductivity.

[1] J.-H. Yu, B.-J. Lee, S. Zhou, J. H. Sung, C. Zhao, C.-H. Shin, B. Yu, G.-L. Xu, K. Amine and J.-S. Yu, ACS Nano **18**, 31974. (2024).

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