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HIGHLIGHT

Dual-functional ion redistributor for dendrite-free lithium metal anodes

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With the rapid development of high-end electronic devices such as electric vehicles and portable electronics products, the currently widely used lithium (Li)-ion batteries are found greatly difficult to meet the growing demand for energy storage [1, 2]. Li metal batteries are highly regarded as a promising alternative because Li metal possesses high theoretical capacity (3860 $mAh \cdot g^{-1}$) and low redox potential (− 3.04 V vs. SHE) [3]. However, the wide applications of Li metal batteries are still hindered by severe challenges. Li dendrite growth, unstable solid electrolyte interphase (SEI), inactive lithium deposition, and large volume change during the repeated plating/stripping process induce low discharging capacity and short cycling life accompanied by safety hazards [4, 5]. To address the aforementioned problems, many effective strategies have been proposed to prevent Li metal anodes from dendrite growth and improve the stability of the anode/electrolyte interface, including adding additives to electrolytes, using safer electrolytes, making artificial SEI, and modifying the anode structure [6–8]. However, these strategies are still unsatisfied. For instance, SEI-stabilized electrolyte additives are easily consumed as the cyclic goes on, the interfacial SEI are insufficiently stable during long-term charge/discharge cycles, and the uneven growth of Li dendrites can hardly be completely hindered in the inner space of three-dimensional (3D) hosts, which greatly limit the development of Li metal batteries. Another prospective route is designing nanoporous structure with uniform lithium-ion flow at the electrolyte/electrode interface, which can fundamentally suppress lithium dendrites and effectively achieve a dendrite-free lithium anode.

Recently, Wu et al. from Dalian Institute of Chemical Physics (Chinese Academy of Sciences, China) reported the hard-template synthesis of a two-dimensional (2D) heterostructure of mesoporous polypyrrole (mPPy) uniformly grown on defective graphene oxide (mPPy-GO) as a new-concept dual-functional Li-ion redistributor (Fig. 1a) [9]. Thanks to the synergistic effect between Li-ion nanotransmission channel of mPPy and Li-ion nanosieves of defective GO, the Li-ion is gradually distributed and uniformly deposited, and a dendrite-free Li metal anode is obtained. This mechanically stable and flexible GO protective films can act as physical barriers to delay the transfer of Li-ions and prevent the "tip effect" due to the intrinsic defect sites of GO (Fig. 1b). In addition, the 2D mPPy-GO heterostructure could act as a dual-functional redistributor to exceptionally dissipate the concentrated Li-ions to realize the uniformization and smooth of Li-ion distribution (Fig. 1c). The mPPy-GO electrode displayed a high Coulombic efficiency (98%) up to 1000 cycles (Fig. 1d), coupled with long cyclability without dendrites growth even under extreme environment (i.e., 0 and 50 °C, Fig. 1e, f). At the low temperature (0 °C), mPPy-GO-Li

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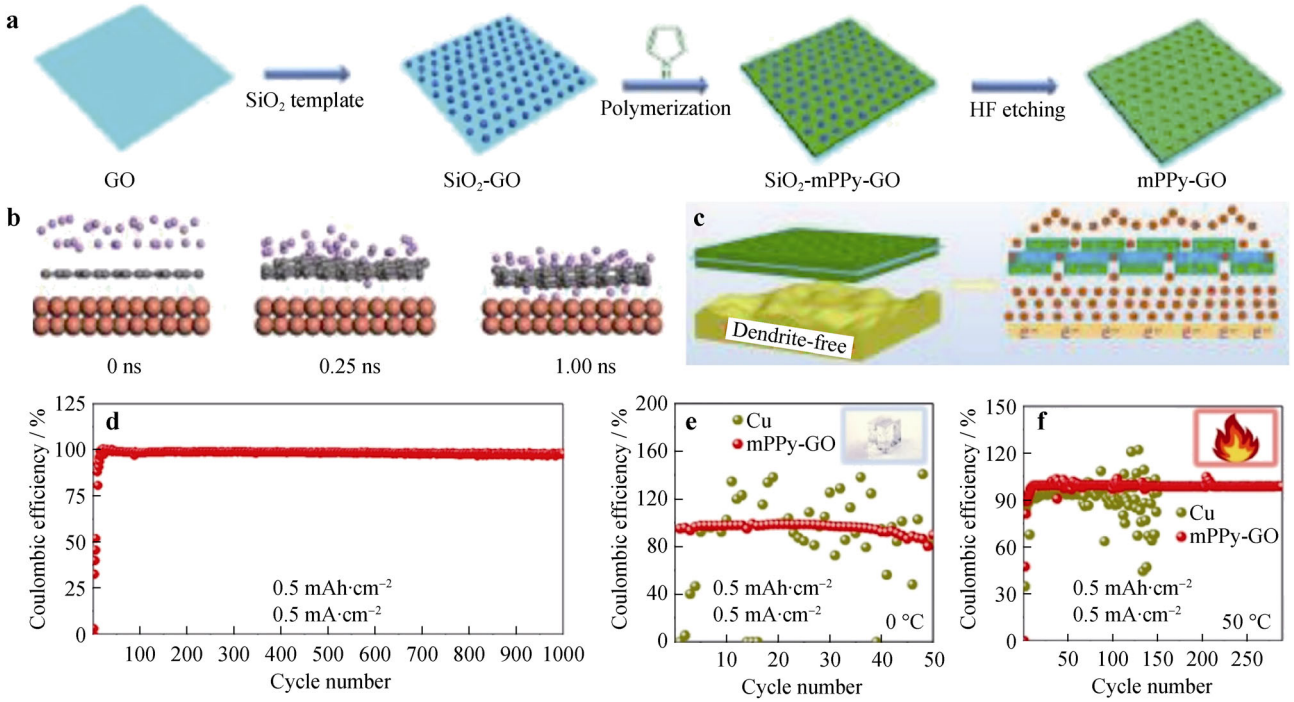

Fig. 1 Schematic illustration and electrochemical characterization of 2D mPPy-GO heterostructure: **a** schematic of fabrication of 2D mPPy-GO heterostructure, **b** atomic configurations of diffusion pathway of Li atoms on Cu current collector with defective GO at different time; **c** schematic illustration of electrochemical deposition behaviors of electrodes with 2D mPPy-GO nanosheets as dual-functional Li-ion redistributor for uniform Li deposition; **d** ultralong time Coulombic efficiency test of mPPy-GO electrode; Coulombic efficiencies of mPPy-GO and Cu electrodes at **e** low temperature of 0 °C and **f** high temperature of 50 °C (Reproduced from Ref. [9]. Copyright 2020, Wiley–VCH)

electrode displayed stable cycle performance more than 50 cycles, while the bare Cu anode revealed deteriorative cycle performance due to the slow electrochemical dynamics (Fig. 1e). Under high temperature of 50 °C, side reactions such as electrolyte decomposition and Li dendrites accelerated the decrease in the cell cycle ability. Surprisingly, the mPPy-GO electrode showed impressively stable Coulombic efficiencies of 98.5% after 290 cycles (Fig. 1f), which clearly demonstrated the unique roles of mPPy-GO in uniformly regulating Li flux to stabilize Li deposition.

This new strategy for solving Li dendrite issues by dual-functional 2D Li-ion redistributor could provide a new paradigm for the design and construction of Li metal anode toward high-energy–density Li batteries. Nevertheless, it is necessary to do more work to further enhance the electrochemical performance of Li metal battery. On the other hand, potential safety issues, relatively low CE, and lack of cost-effective scalable manufacturing required to be promoted are investigated for practical commercial applications.

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