High rate and stable cycling of lithium metal anode

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Battery energy storage system has achieved great success in the portable electronics and electronic vehicles. Among various battery electrode materials, lithium metal is regarded as the “Holy Grail” electrode due to the extra-high capacity (3860 mAh g⁻¹), the lowest negative electrochemical potential (−3.040 V vs. the standard hydrogen electrode) and a small gravimetric density of 0.534 g cm⁻³. However, unstable solid electrolyte interphase (SEI) and lithium dendrite growth during lithium plating/stripping induce poor safety, low coulombic efficiency, and short span life of lithium metal batteries. These problems have hindered the commercialization of the secondary lithium batteries for decades.

Recently, direct surface modification on Li metal was reported to be an effective strategy for improving the rate capability and cycling stability of the Li metal by increasing its surface area and making some patterns on a piece of smooth Li foil. The above success demonstrates the feasibility to change the plating and stripping behaviors of lithium by modifying the chemical properties of the electrode surface. However, their limitations, such as the complex synthesis methods and high cost, still restrict the commercialization of Li metal and demonstrate that the state-of-the-art knowledge is inadequate and more effort is required to understand the growth mechanism of the Li dendrites.

Herein, we try a facile preparation to form a uniform SEI. We developed a conformal LiF surface coating strategy enabled by the reaction between fluoride and Li metal. It is proved that the cycle stability of the lithium cells can be remarkably prolonged, because of the compact and homogeneous SEI layers consisting of LiF products formed on the lithium metal surface. Symmetric cell cycling corroborated the enhanced stability with LiF coating in prolonged cycles, during which neither voltage fluctuation nor increase could be observed. Moreover, Li-LTO prototype cells with LiF-coated layered Li as the anode were examined, which exhibited further improved cycling stability and confirmed good surface passivation and thus reduced shuttle effect. By realizing high quality of LiF coating on any forms of Li surface, this work demonstrates the great feasibility as well as the advantages of fluoride reactions for Li surface protection, and it paves the way for the stable and safe operation of Li metal anode in the future. The as-developed methodology can also inspire further efforts on Li surface.