New artificial interphase with Li-ion pathway between Li metal and electrolytes for secure lithium metal anodes

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While Li-ion batteries were firstly studied with Li metal as anodes due to its high theoretical specific capacity (3860 mA h g^{-1}) and lowest negative electrochemical potential (-3.040 V vs SHE), commercial Li-ion batteries since 1992 consisted of graphite as an anode due to critical safety issues. However, for several decades, many strategies have been developed to surmount difficulties like problems of dendrite growth using modifications of electrolytes with additives, a change of lithium metal morphology, and an introduction of chemical/physical coating layers on Li metal.

Herein, we report a simple approach to improve the electrochemical performance of Li metal anodes in Li-ion batteries with commercial carbonate-based electrolytes by utilizing a high Li-ion conductor, Li_2TiO_3, on Li metal anodes. Monoclinic Li_2TiO_3 is electrochemically inert in a wide voltage range with excellent structural stability in a liquid electrolyte. In addition, it structurally has a 3D path for Li-ion diffusions, leading to more uniform Li-ion flux. Li_2TiO_3 nanoparticles with a size of sub-100 nm were synthesized via a sol-gel method, and then the resulting particles were mixed with carbon as a conducting material and PVDF-HFP as a binder. A bright Li metal surface was covered with gray Li_2TiO_3 films. From various Li plating and striping tests with pristine and modified Li metal as working electrodes, the modified Li metal exhibited outstanding electrochemical performances such as stable coulombic efficiency over 100^{th} cycles at 1 mA/cm^2. Moreover, the modified Li metal was applied for practical lithium-ion batteries as a counter electrode to confirm the protective effects with LMO as a working electrode at a room temperature (25 ^oC) and high temperature (60 ^oC).