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Garnet-based Bilayer Hybrid Solid Electrolyte for High-Voltage Cathode Material Modified with Composite Interface Enabler on Lithium-Metal Batteries

Authors : Kumlachew Zelalem Walle, Chun-Chen Yang

Abstract : Solid-state lithium metal batteries (SSLMBs) are considered promising candidates for next-generation energy storage devices due to their superior energy density and excellent safety. However, recent findings have shown that the formation of lithium (Li) dendrites in SSLMBs still exhibits a terrible growth ability, which makes the development of SSLMBs have to face the challenges posed by the Li dendrite problem. In this work, an inorganic/organic mixture coating material (g-C3N4/ZIF-8/PVDF) was used to modify the surface of lithium metal anode (LMA). Then the modified LMA (denoted as g-C_3N_4@Li) was assembled with lithium nafion (LiNf) coated commercial NCM811 (LiNf@NCM811) using a bilayer hybrid solid electrolyte (Bi-HSE) that incorporated 20 wt.% (vs. polymer) LiNf coated Li6.05Ga0.25La3Zr2O11.8F0.2 (LiNf@LG0.25LZOF) filler faced to the positive electrode and the other layer with 80 wt.% (vs. polymer) filler content faced to the g-C_3N_4@Li. The garnet-type Li6.05Ga0.25La3Zr2O11.8F0.2 (LG0.25LZOF) solid electrolyte was prepared via co-precipitation reaction process from Taylor flow reactor and modified using lithium nafion (LiNf), a Li-ion conducting polymer. The Bi-HSE exhibited high ionic conductivity of 6.8 □ 10–4 S cm–1 at room temperature, and a wide electrochemical window (0–5.0 V vs. Li/Li+). The coin cell was charged between 2.8 to 4.5 V at 0.2C and delivered an initial specific discharge capacity of 194.3 mAh g–1 and after 100 cycles it maintained 81.8% of its initial capacity at room temperature. The presence of a nano-sheet g-C3N4/ZIF-8/PVDF as a composite coating material on the LMA surface suppress the dendrite growth and enhance the compatibility as well as the interfacial contact between anode/electrolyte membrane. The g-C3N4@Li symmetrical cells incorporating this hybrid electrolyte possessed excellent interfacial stability over 1000 h at 0.1 mA cm–2 and a high critical current density (1 mA cm–2). Moreover, the in-situ formation of Li3N on the solid electrolyte interface (SEI) layer as depicted from the XPS result also improves the ionic conductivity and interface contact during the charge/discharge process. Therefore, these novel multi-layered fabrication strategies of hybrid/composite solid electrolyte membranes and modification of the LMA surface using mixed coating materials have potential applications in the preparation of highly safe high-voltage cathodes for SSLMBs.

Keywords : high-voltage cathodes, hybrid solid electrolytes, garnet, graphitic-carbon nitride (g-C3N4), ZIF-8 MOF
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