

Minimizing Ion/Electron Pathways Through Ultrathin Conformal Holey Graphene Encapsulation in Li- and Mn-Rich Layered Oxide Cathodes for High-Performance Lithium-Ion Batteries

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Lithium-ion batteries (LIBs) are increasingly favored due to their attractive features. The design of the cathode in LIBs plays a critical role in determining cell capacity, operating voltage, and overall cost. Lithium- and manganese-rich (LMR) cathode materials stand out as promising candidates for the next generation of cathode materials due to their ability to operate at high voltages and provide capacity exceeding 250 mAh g⁻¹. However, despite these appealing characteristics, LMR faces challenges in commercialization due to factors such as poor rate capability and rapid capacity and voltage decay during cycling. These are closely associated with the fundamental structural issues arising from the two distinct phases of LMR materials, slow reaction kinetics and structural degradation occurring through side reactions between the electrode and electrolyte.

In this study, we introduce a carbon encapsulation technique that integrates polyethylenimine (PEI) and holey graphene onto the LMR surface, aiming not only to augment electrical conductivity but also to facilitate ionic conductivity. Despite its low carbon content of 0.1 wt%, the suggested PEI/holey graphene-encapsulated LMR demonstrates enhanced cycle stability and rate performance for the LMR electrode. Moreover, the thin and uniform PEI/holey graphene encapsulation layer serves a dual purpose by easing the movement of Li⁺/e⁻ on the LMR surface and providing a protective barrier against physical and chemical aggressions. Throughout cycling assessments, the PEI/holey graphene-encapsulated LMR mitigates the leaching of transition metals, mitigating microcrack formation and irreversible structural alterations compared to bare LMR. Consequently, the proposed PEI/holey graphene encapsulation emerges as an attractive technology for high-performance LIB design, concurrently elevating the cycle stability and rate performance of LMR electrodes.

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