Hierarchical Functional Layers on High-Capacity Lithium-Excess Cathodes for Superior Lithium Ion Batteries

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Abstract: The Li-excess layered cathode material, $Li[Li_{0.2}Mn_{0.54}Ni_{0.13}Co_{0.13}]O_2$ (LMNCO), has attracted tremendous attention as a promising candidate for high-energy and high-power lithium ion batteries due to its large theoretic capacity. LMNCO nanoparticles are facilely synthesized via using surfactant-assisted dispersion in this work. In order to improve cycleability and rate capability of LMNCO cathode, ultrathin and conformal oxide coatings are deposited on the surface of individual LMNCO nanoparticle via atomic layer deposition (ALD). The effect of oxide ALD coatings on improving electrochemical performance of LMNCO nanoparticles is evaluated and optimized via tuning the coating thickness and composition. In addition, we synthesize a novel core-shell structure cathode consisting of Li-stoichiometric material as shell and Li-excess LMNCO as core, and its electrochemical property is optimized by tailoring weight content and composition of shell material. Finally, electrochemical performance of Li-excess cathode material can be maximized by surface modification with hierarch functional layers composed of $LiCoO_2$ shell and ZrO_2 ALD coating. LMNCO nanoparticles modified with 10 wt.% $LiCoO_2$ shell (~10 nm thick) and six ZrO_2 ALD layers (~1 nm thick) delivers the best performance, with initial discharge capacities of 296.4, 259.8, 156.6 and 104.2 mAh g^{-1} at 0.1*C*, 1*C*, 5*C* and 10*C*, and can retain 184.0 mAh g^{-1} at 1*C* after 100 electrochemical cycles, which is almost three times the retained capacity of bare LMNCO cathode. Such remarkable reversibility, cycleabilitiy and rate capability of nanoarchitected Li-excess layered cathode material can be attributed to the synergic effect from hierarch functional coatings to reduce electrochemical polarization, structural degradation and side reactions during electrochemical cycling.

Keywords: Li-excess layered cathode material; atomic layer deposition; core-shell structure; hierarchical functional layers; lithium ion battery