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Effect of Li-excess on Electrochemical Performance of Ni-rich $LiNi_{0.9}Co_{0.09}Mn_{0.09}O_2$ Cathode Materials for Li-ion Batteries

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Abstract : Nickel-rich layered oxide cathode materials having a Ni content of ≥ 90% have great potential for use in next-generation lithium-ion batteries (LIBs), due to their high energy densities and relatively low cost. They suffer, however, from poor cycling performance and rate capability, significantly hampering their widespread applicability. In this study we synthesized a Ni-rich precursor through a co-precipitation method and added different amounts of Li-excess on the precursors using a solid-state method to obtain sintered Li1+x(Ni0.9Co0.05Mn0.05)1–xO2 (denoted as L1+x-NCM; x = 0.00, 0.02, 0.04, 0.06, and 0.08) transition metal (TM) oxide cathode materials. The L1+x-NCM cathode having a Li-excess of 4% exhibited a discharge capacity of ca. 216.17 mAh g–1 at 2.7–4.3 V, 0.1C and retained 95.7% of its initial discharge capacity (ca. 181.39 mAh g–1) after 100 cycles of 1C charge/discharge which is the best performance as compared with stoichiometric Li1+x(Ni0.9Co0.05Mn0.05)1-xO2 (i.e. x=0, Li:TM = 1:1). Furthermore, a high-rate capability of ca. 162.92 mAh g–1 at a rate of 10C, led to the 4% Li-excess optimizing the electrochemical performance, relative to the other Li-excess samples. Ex/in-situ X-ray diffraction, scanning electron microscopy, and X-ray photoelectron spectroscopy revealed that the 4% Li-excess in the Ni-rich NCM90 cathode material: (i). decreased the Li+/Ni2+ disorder by increasing the content of Ni3+ in the TM slab, (ii). increased the crystallinity, and (iii). accelerated Li+ ion transport by widening the Li-slab. Furthermore, electrochemical impedance spectroscopy and cyclic voltammetry confirmed that the appropriate Li-excess lowered the electrochemical impedance and improved the reversibility of the electrochemical reaction. Therefore, our results revealed that NCM90 cathode materials featuring an optimal Li-excess are potential candidates for use in next-generation Li-ion batteries.

Keywords : $LiNi_{0.9}Co_{0.09}Mn_{0.09}O_2$, li-excess, cation mixing, structure change, cycle stability, electrochemical properties
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