Electrochemical and Interfacial Reaction Behavior of High-Voltage LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ Cathode for Lithium-ion Batteries

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Multi-component cathode material of $\text{LiNi}_{1-x-y}\text{Co}_x\text{Mn}_y\text{O}_2$ with layered hexagonal structure has been regarded as a promising cathode material due to lower cost, less toxicity, higher thermal stability and higher operation voltage than the single-component materials.¹ Its performance is however often difficult to be realized, in particular, at high voltage operation (> 4.3 V vs. Li/Li⁺), due to severe oxidative decomposition of conventional electrolyte. Understanding the cathode-electrolyte interfacial reaction at high voltage and improving the high voltage stability are highly demanded to obtain a stable cycling performance.^{2,3} Here we report a new electrolyte formulation, which withstands at 4.6 V, and its application to the lithium cell with LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ cathode. Correlation of cathode-electrolyte interfacial reaction behavior and cycling ability is discussed.

Cathode active material of LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ was synthesized at 900 °C in air using the coprecipitate precursor of Ni_{0.5}Co_{0.2}Mn_{0.3}(OH)₂ (EMT). Composition was determined by energy dispersive X-ray spectroscopy. The crystal structure of coprecipitate precursor and cathode material, and the absence of impurity were identified by X-ray diffraction analysis, measured in the 20 range of 10 - 80° with the scan rate of 2°/min. Lithium coin cells, consisted of LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ as a working electrode, a lithium foil as counter electrode, PE separator and electrolyte of 1M LiPF₆/EC:EMC with 5 wt% additive was assembled in the Ar-filled glove box. The cells were evaluated for their cycling ability at C/10 rate between 3.0 and 4.6 V.

Fig. 1 shows capacity vs. voltage plots obtained at the 1^{st} , 10^{th} and 30^{th} cycle of LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ cathode with additive. The initial charge and discharge capacities are 276 and 205 mAh/g, respectively, with initial coulombic efficiency of 74 %. After 30 cycles, the cathode exhibits capacity retention of 88 % with the discharge capacity of 188 mAh/g, in contrast to a rapid capacity fade without additive. The use of additive is effective in improving high voltage stability and cycling ability of LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ cathode. Further discussion of formation and stability of solid electrolyte interface (SEI) would be presented in the meeting.

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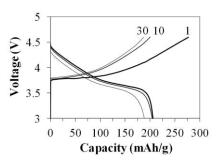


Fig. 1. Capacity vs. voltage plots of $Li(Ni_{0.5}Co_{0.2}Mn_{0.3})O_2$ cathode with electrolyte additive at C/10.