Investigation of metal dissolution of LiNi$_{0.5}$Mn$_{1.5}$O$_4$ based cathode materials in lithium ion batteries

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Abstract

Lithium-ion batteries (LIBs) are currently used as the dominant energy sources for large-scale applications (hybrid electric vehicles (HEV) and electric vehicles (EV)) and portable electronic devices owing to their high energy density, low self-discharge, and long cycle life. High operating potential (~4.7 V vs. Li) of spinel LiNi$_{0.5}$Mn$_{1.5}$O$_4$ (LNMO) is believed one of the most promising cathode materials for Li-ion batteries. The intrinsically fast Li$^+$ diffusion in the three-dimensional spinel structured cathode materials demonstrate the great rate capability and cycling stability. It is well known that spinel LNMO cathodes can exist in the form of two crystalline structure, either in an ordered (P4$_3$3$_2$) or a disordered (Fd3m) structure. Although, high-voltage spinel LNMO suffering due to its limited cycle life, especially at higher temperature, which is triggered by decomposition of electrolyte or dissolution of Mn/Ni occurs at high operation potential. The Fd3m space group possesses the faster Li$^+$ ion diffusion ability than that of P4$_3$3$_2$ space group in the crystalline structure. The calcination temperature plays a vital role in the determination of the surface morphology and crystalline structures (Fd3m and P4$_3$3$_2$). Thus, by controlling and varying the calcination temperatures of the LNMO materials can easily alter the electrochemical performance.

LNMO cathode materials with a spinel structure were synthesized by sol-gel method at various calcination temperature ranges from 600 to 1000 °C. The physical characterization (XRD, PSA, TGA, RAMAN), surface morphology (FE-SEM) and electrochemical evaluations (CV, EIS and galvanostatic charge–discharge) revealed that the effectiveness of the calcination temperatures on enhancing the electrochemical performances of the LNMO electrode materials. In addition, dissolution test was performed to estimate the amount of metal ion dissolutions to explore the capacity fading mechanism of LNMO electrode materials. The structural analysis exhibited that the controlling the calcination temperature contributes to the high crystalline nature and space group changes. The enhanced electrochemical performance was observed at the calcination temperature of 900 °C owing to the presence of higher proportion of the Fd3m space group than that of P4$_3$3$_2$ in the crystalline LNMO structure.

Key Words: Spinel LiNi$_{0.5}$Mn$_{1.5}$O$_4$, sol-gel, Cathode, lithium-ion batteries