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Interplay between Two-Phase and Solid Solution Reactions in High Voltage Spinel Cathode Material for Lithium Ion Batteries

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Abstract

Lithium ion batteries (LIBs) are attracting intensive interests worldwide because of their potential applications in transportation electrification and utility grid. The intercalation compounds used in LIBs electrochemically react with Li^+ ions via single or multiple phase transitions depending on the nature of the material structure as well as the synthesis and operating conditions. For $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ high voltage spinel, a promising candidate positive electrode material for LIBs, there are three spinel-structured phases sequentially appeared through two successive two-phase reactions during the delithiation/lithiation processes. Here we demonstrate, experimentally and theoretically, that through elemental substitution, the solid solution ranges for both the first and second phases are significantly extended during the electrochemical charge-discharge process. This type of structural changes with more solid solution regions facilitate fast Li^+ diffusion by reducing the number of phase boundaries that Li^+ ions have to overcome and resulted in less shrinkage of the unit cells at the end of charge process. This work unravels the fundamental interactions between structural and electrochemical properties by using spinel as the platform, which may be widely adopted to explain or tailor the properties of materials for energy storage and conversion.

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