Li-Removal Mechanism and Its Effect on Oxygen Stability Influencing the Electrochemical Performance of $Li_{1.17}Ni_{0.17}Mn_{0.67}O_2$: Experimental and First-Principles Analysis

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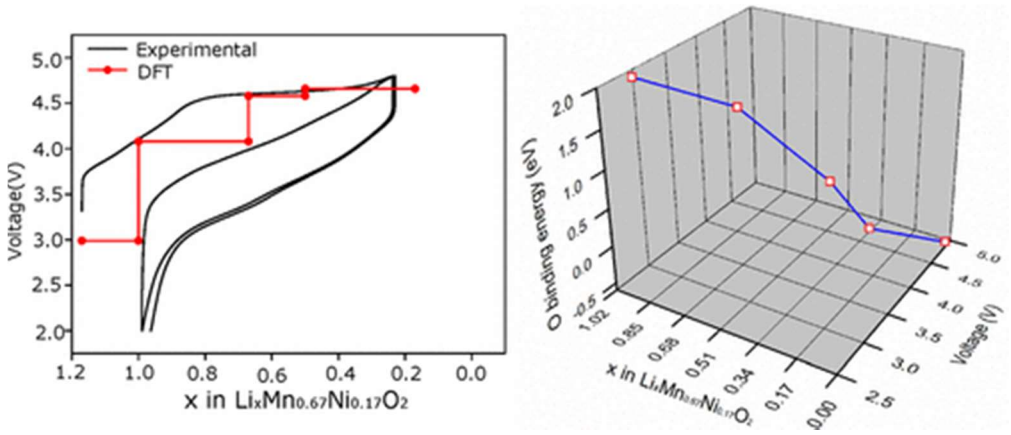
Abstract



The high capacity of Li-rich layered cathode materials is always accompanied by the removal of oxygen from the crystal structure. These oxygen vacancies alter the structural stability, which subsequently deteriorates the electrochemical performance. The electronic origin of oxygen stability with partial delithiation has not been extensively studied so far in the presence of multiple d-orbital elements. Current work presents the experimental and density functional theory based study of the Li-rich phase, $Li_{1.17}Ni_{0.17}Mn_{0.67}O_2$. This study reveals the lithium removal mechanism and its influence on the oxygen stability. Further, the study suggests how lithium removal from different lithium sites, i.e., 2b, 2c, and 4h Wyckoff positions, influence the partial intercalation potential. On higher degree of delithiation, electrochemical potential increases and oxygen binding energy decreases. Thus, the oxygen stability reduces in the compound. At this stage, the material becomes metallic with zero band gap, which facilitates oxygen loss. This affectively influences the charge transfer process and redox center of the compound, which has been captured in this study.