

Oxygen stability study of Li-rich compound: First-Principles Analysis

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ABSTRACT

High energy density Li-rich layered cathode materials suffer from structural instability at high voltage. It is known that oxygen stability influence the structural stability of Li_2MO_3 ($\text{M}=\text{Co}/\text{Mn}/\text{Ni}$). Oxygen stability with partial de-lithiation has not been clarified in presence of multiple d-orbital elements. This work presents density functional theory based study of $\text{Li}_{1.17}\text{Ni}_{0.17}\text{Mn}_{0.67}\text{O}_2$. In the series of Li-rich compounds, end point material is Li_2MnO_3 , in which oxidation of Mn^{4+} compensates with oxidation of O^{2-} to O_2 while charging. This makes Li_2MnO_3 electrochemically inactive poor cathode material. Li_2MnO_3 become electro-chemically active when it is stabilized with LiMO_2 . In Li-rich compound, Ni acts as a structurally stabilizing agent and it prevents oxygen evolution during cycling. On higher degree of de-lithiation, oxygen binding energy decreases. At this stage material becomes metallic with zero band gap which facilitates oxygen loss. Lithium removal from the metal layer and lithium layer influence the oxygen binding energy, which is also related with the structural stability. The main aim of this work is to study oxygen stability of the material.