

**Exploring anionic redox reactions of non-overlithiated layered oxides:
LiNi_{1/3}Co_{1/3}Mn_{1/3}O₂**

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In recent years, an interesting charge compensation mechanism by anions, also called anionic redox, has attracted a lot of attention in lithium ion battery society because it has considered as promising strategy to overcome the capacity limitations of classical cathode materials. Another important implication of this research trend is that it triggered the interest on anionic contribution in charge compensation of cathode materials. However, recent studies dealing with anionic redox are all concentrated on overlithiated materials. Here, we tried to unveil the anionic redox behavior in LiNi_{1/3}Co_{1/3}Mn_{1/3}O₂ (NCM111), one of the representative layered oxide cathode materials, by reexamining the charge compensation mechanism of NCM111. Even though charge compensation mechanism of NCM111 in the practical voltage range (< 4.3 V) have revealed in detailed by a variety of studies, the charge compensation mechanism in the high voltage region still remains unclear. Finally, through the detailed analysis, it is confirmed that the oxygen in the metal-oxygen bond contributes to the charge compensation in the low-voltage range, and the lone-pair oxygen participates in charge compensation in the high-voltage range. This finding is expected to not only provide a new perspective for anion redox researchers but also help establish a high voltage stabilization strategy for layered oxide materials.