**Anionic redox reactivity and high-energy-density cathode materials for Li-ion battery**

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**Abstract:** Lithium ion battery (LIB) has been the most prominent electrochemical energy storage technology over the past decades and enabled the wireless evolution of portable electronic devices. The expanded use of renewable but intermittent energy sources coupled with increasing demand for electric transportation vehicles, put forward requirements to LIBs for higher capacity, lower cost, and longer cycle life. Therefore, the development of new high energy electrode materials for rechargeable lithium ion batteries is becoming more essential. State-of-the-art electrodes like LiCoO2 and LiMn2O4 function *via* the intercalation mechanism during Li accommodation and extraction with the preserved crystal structure, accompanied by redox reactions of transition metal cations. The specific capacity of the electrode is then limited by the number of electrons per transition metal cation that can participate in the redox reaction. The exclusive transition metal redox is challenged by the recent discovery of anionic redox reactivity in Li-excess materials, which can be taken advantage of to greatly boost the capacity of anionic redox active electrodes. The density functional theory (DFT) has been widely used as effective tools to study battery materials to explore new, high-performance electrode materials, and understand in detail the underlying mechanisms during electrochemical reactions. Using DFT calculations, we reveal the anionic redox electrochemistry by examining the oxidation state evolution along with the local atomistic environment variation and identify the origin of the anionic redox reactivity. Armed with this knowledge, we further predict new electrode materials with improved electrochemical properties through high-throughput DFT calculations.