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Phase Stability and Electro-Mechanical Properties of LiMn1.5Ni0.5O4 via Cluster Expansion and DFT

High-voltage spinel LiMn1.5Ni0.5O4 (LMNO) is a promising material for next-generation lithium batteries. It has attracted interest as a high-voltage cathode due to its theoretical operating potential of 4.7-4.8 V versus Li^+/Li. However, its practical implementation faces substantial challenges from structural degradation and capacity fade during cycling, originating from complex phase transformations and electronic structure changes during de-lithiation. The fundamental mechanisms governing these degradation processes remain incompletely understood, particularly regarding the interplay between Li/vacancy ordering, transition metal redox chemistry, and lattice stability. This computational study employs cluster expansion and density functional theory (PBEsol+U/HSE06) to systematically investigate the de-lithiation pathway in LiMn1.5Ni0.5O4 (x = 1⟶0). We aim to establish structure-property relationships by examining: (1) phase stability evolution, (2) redox behaviour of Ni and Mn cations, (3) mechanical response to lithium extraction, and (4) thermodynamic voltage profiles of the bulk material. Our calculations reveal sequential oxidation processes (Ni^{2+}⟶Ni^{4+} preceding Mn^{3+}⟶Mn^{4+}) accompanied by Jahn-Teller distortions and identify critical de-lithiation thresholds (x < 0.25) where oxygen lattice destabilization occurs. The material maintains remarkable mechanical stability (bulk modulus >120 GPa throughout de-lithiation), suggesting structural resilience despite electronic structure modifications. These findings provide fundamental insights into LMNO's degradation mechanisms while establishing a computational framework for evaluating stabilization strategies.

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