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Tailoring structural design of spinel LiMn2O4 cathode material through high entropy doping

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The spinel LiMn2O4 cathode material has attractive candidates for the design and engineering of cost-effective and thermally sustainable lithium-ion batteries for optimal utilisation in electric vehicles and smart grid technologies. Despite its electrochemical qualities, its commercialization is delayed by the widely reported capacity loss during battery operation. The capacity attenuation is linked to structural degradation caused by Jahn-Teller active and disproportionation of Mn3+ ions. In several studies, the structural stability of spinel LiMn2O4 was improved by single- or dual-doping the Mn sites to curtail the number of Mn3+ ions. However, this results in loss of active ions, which ultimately limits the amount of energy that can be obtained from the battery. Herein, a high-entropy (HE) doping strategy is used to enhance the structural stability and electrochemical performance of LiMn2O4 spinel. The unique interactions of various dopants in HE doping yield enhanced structural stability and redox coupling, which can improve the concentration of the active material in the system. An HE-doped LiMn2O4 (LiMn1.92Mg0.02Cr0.02Al0.02Co0.02Ni0.02O4) spinel structure was successfully optimized using the Vienna Ab initio Simulation Package (VASP) code. The lattice parameters of the optimized (ground state) structure were determined to be 8.270 Å, which is less than the value of 8.274 Å of the pristine LiMn2O4 spinel structure. The yielded lattice contractions suggest a stronger M-O bond beneficial for increased resistance to phase changes and degradation. Moreover, the concentration of Mn3+ was decreased by 5.3% to defer the onset of the Jahn-Teller distortion and enhance capacity retention. This retention is part of some significant benefits emanating from dopants such as Cr3+ as it can participate in storing electric charge during the charging process by forming Cr4+ thus compensating the capacity loss endured during Mn3+ concentration reduction. Consequently, this work paves a path for exploration of several other fundamental properties linked to the electrochemical performance of spinel.

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