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Zr-Doping of P2-NaxMn0.625Ni0.375O2-Layered Oxides for High-Energy Na-Ion Battery Cathodes: New Insights from First-Principles

Na-ion batteries (NIBs) are at the forefront of energy storage technology due to larger abundance of raw materials and enhanced cost-effectiveness compared to Li-ion counterparts [1]. Despite the great advances achieved in last decades, identification of efficient, stable and cheap component materials still hinders a convenient NIB commercialization. Following the route paved by different TM combinations in P2-cathode materials [2], we present a first-principles investigation at the PBE + U(-D3BJ) level of theory on Zr-containing NaxTMO2 (TM = Zr, Ni, and Mn), chosen for its potential to enhance structural stability and electronic properties [3,4]. We dissect the specific roles of different element sublattices during sodiation/desodiation processes. In addition, oxygen vacancies and TM anti-sites defects are addressed in the high-voltage range (%Na = 25% ⍰ 12.5%), and their related formation energies and both structural and electronic features are discussed. Experimental analysis (e.g. toward cyclic voltammetry and X-rays diffraction) provide key validation for computational results demonstrating the cathode stability and efficiency. All the theoretical insights and the validating experimental results can be useful for an optimal design for next-generations efficient and high-energy NIB cathodes with enhanced structural stability at high voltage and improved device sustainability.

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