Enhanced High-Voltage Cyclability of O3-Type Sodium Layered Oxide Cathode through Ca-substitution

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To facilitate the practical realization of sodium-ion batteries(SIBs), the energy density, determined by the output operating voltage and/or capacity, needs to be improved to the level of commercial Li-ion batteries(LIBs). To date, O3-type $Na[Ni_{0.5}Mn_{0.5}]O_2$ cathode has been considered as one of the most promising cathode materials because of its high capacity and good practical applicability. Although an effective approach would be to elevate the operating voltage of the battery to reach higher energy density of SIBs, however, O3-type $Na[Ni_{0.5}Mn_{0.5}]O_2$ generally experiences irreversible phase transition occurs via the redox reaction of Ni^{2+}/Ni^{4+}, which affects the cycling stability within the high-voltage window. In this study, we introduced the Ca substitution into bulk Na sites, which significantly improved the structural and electrochemical properties of the Na $[Ni_{0.5}Mn_{0.5}]O_2$ cathode. The presence of Ca^{2+} ions in Na sites sufficiently stabilized the crystal structure and effectively mitigated the volume expansion when Na^+ ions are almost fully extracted from the structure. Moreover, Ca substitution improves the practically useful aspects such as thermal and air stability.