

# Na-doped Method of Layered $\text{Li}_{1.2}\text{Mn}_{0.54}\text{Ni}_{0.13}\text{Co}_{0.13}\text{O}_2$ for High Capacity and High-rate Li-ion Batteries

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High energy and power-density lithium-ion batteries (LIBs) are critically needed for applications in aerospace, EVs, energy storage systems and so on<sup>[1]</sup>. Lack of high performance cathode materials has become a technological bottleneck for the commercial LIBs. Due to the high reversible capacity (300mAh/g), layered  $\text{Li}_{1.2}\text{Mn}_{0.54}\text{Ni}_{0.13}\text{Co}_{0.13}\text{O}_2$  has been paid much attention<sup>[2]</sup>. Despite its high specific capacity,  $\text{Li}_{1.2}\text{Mn}_{0.54}\text{Ni}_{0.13}\text{Co}_{0.13}\text{O}_2$  suffers poor rate capability, which was ascribed to two-dimensional ion channel caused by layered structure. Moreover, layered  $\text{Li}_{1.2}\text{Mn}_{0.54}\text{Ni}_{0.13}\text{Co}_{0.13}\text{O}_2$  has irreversible structural rearrangement above 4.5V during the first charge process. The irreversible capacity loss is due to the oxidation of  $\text{O}^{2-}$  ions, which involves the extraction of lithium as  $\text{Li}_2\text{O}$  during first charge process and the inability to insert part of the extracted lithium back into the lattice during the first discharge process<sup>[3]</sup>. Many efforts have been devoted to address these challenges.

Here we report a reasonable design and synthesis of cathode material. Na-doped  $\text{Li}_{1.2}\text{Mn}_{0.54}\text{Ni}_{0.13}\text{Co}_{0.13}\text{O}_2$  has been synthesized by high-temperature solid-state molten salt method for the first time. Our results indicate that  $\text{Na}^+$  ion enters the lattice instead of the  $\text{Li}^+$  ion with no change the structure. Compared to pristine sample, Na-doped sample has excellent rate and capacity performance, and the irreversible capacity loss decreases from 75 to 40mAh/g. The performance improvements are responsible for the larger ion radius of  $\text{Na}^+$  ion (102pm) than that of  $\text{Li}^+$  ion (76pm), as shown in Fig.1. Due to a proper amount of sodium is substituted for the position of lithium, the  $\text{Li}^+$  ion diffusion channel of the 2D layered structure is expanded, which is beneficial to the lithium ions insertion/extraction reaction. At the same time, the oxygen vacancy can be reduced above 4.5V, which reduces the irreversible loss and improves the stability of the material.

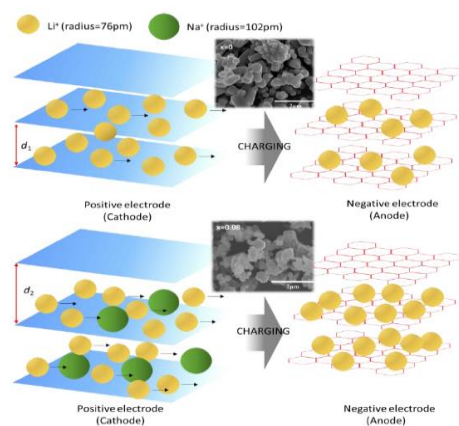


Fig.1 Schematic of Na doping in  $\text{Li}_{1.2}\text{Mn}_{0.54}\text{Ni}_{0.13}\text{Co}_{0.13}\text{O}_2$  material.

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