

# Layer-structured P2-Na<sub>0.66</sub>[Mn<sub>0.6</sub>Ni<sub>0.4-x</sub>Mg<sub>x</sub>]O<sub>2</sub> Cathode Materials for Sodium-ion Batteries

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Sodium-ion batteries (SIBs) have been considered as a promising alternative for large scale energy storage application due to the nature abundance and low cost of sodium. However, the lack of high performance cathode materials is one of the major obstacles for developing high performance room temperature SIBs. Over the past years, various of cathode materials have been investigated, including layered oxides, tunnel-structured oxides, polyanion compounds, and organic compounds. Among them, layered oxides are promising cathode materials with a general formula of Na<sub>x</sub>TMO<sub>2</sub> (TM=transition metals), which can be categorized into two main groups: P2-type and O3-type. P2 and O3 are defined according to the oxygen stacking sequence and coordination environment of the alkali ions. Generally, O3-type compounds have higher reversible capacity compared with P2-type compounds, but their cycling stability is not good and many of them are not stable in air. In contrast, P2-type layered cathode materials usually deliver better cycle performance and rate capability due to the large trigonal prismatic sites occupied by Na<sup>+</sup> which is beneficial for Na-ion transportation.

Here, we present a Mg-substituted layer-structured P2-Na<sub>0.66</sub>[Mn<sub>0.6</sub>Ni<sub>0.4-x</sub>Mg<sub>x</sub>]O<sub>2</sub> ( $0 < x < 0.2$ ) cathode material for sodium-ion batteries. It is found that the lattice parameter *c* increases with the increase of *x* value from 0 to 0.1, then decreases with the increase of *x* value from 0.1 to 0.2, suggesting that the substituted sites change from transition metal site to sodium site with the increase of *x* value (critical value:  $x = 0.1$ ). By using synchrotron-based X-ray diffraction technique, it is revealed that the Mg ions in sodium sites can serve as “pillar” to prevent the collapse of the layered structure in the *c* direction during high voltage charging (suppressing the formation of O2 phase). The stable structure of P2-Na<sub>0.66</sub>[Mn<sub>0.6</sub>Ni<sub>0.2</sub>Mg<sub>0.2</sub>]O<sub>2</sub> ensures a high reversible capacity of 130 mAh g<sup>-1</sup> with excellent rate capability (Figure 1). The charge compensation mechanism and rate-dependent structure change of P2-Na<sub>0.66</sub>[Mn<sub>0.6</sub>Ni<sub>0.2</sub>Mg<sub>0.2</sub>]O<sub>2</sub> were also studied and unveiled.

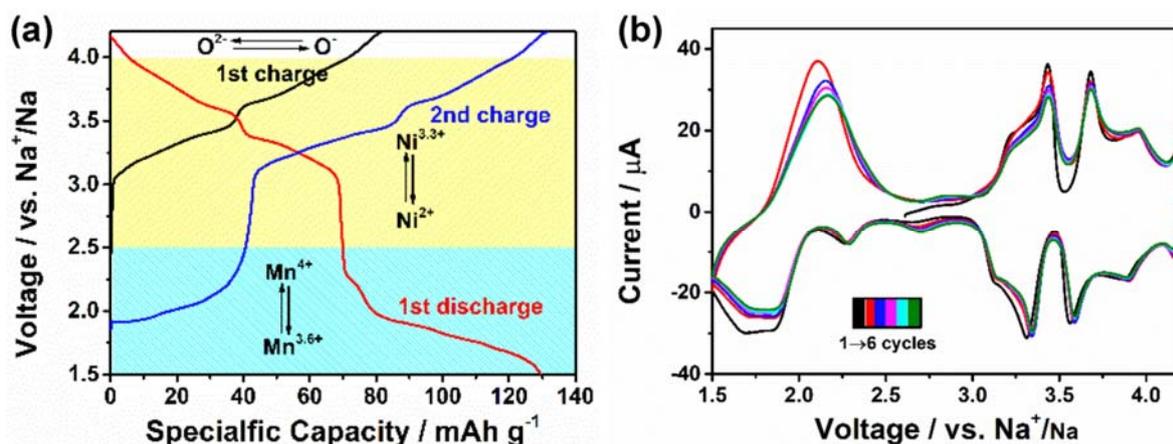


Figure 1 Charge-discharge (a) and cyclic voltammogram (b) profiles of Na<sub>0.66</sub>[Mn<sub>0.6</sub>Ni<sub>0.4-x</sub>Mg<sub>x</sub>]O<sub>2</sub> cathode in Na cells.