

# Promising Candidates for Lithium Ion Batteries: High Nickel Lithium-Rich Layered Cathode Nanomaterial

Weiguo Chu

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Over the past decade, much attention has been paid to the development of lithium-ion batteries (LIBs) owing to the increasing demand for power sources with higher energy and power density. LiCoO<sub>2</sub> has been one of the most widely used cathode materials for its easy synthesis and excellent reversibility since the commercialization of LIBs. However, LiCoO<sub>2</sub> also suffers from many drawbacks such as high cost, toxicity and relatively low practical capacity (about 140 mAh g<sup>-1</sup>, around half of its theoretical capacity), which limits its extensive application [1,2]. Therefore, new types of alternative materials are quite urgent as far as practical applications are concerned. Recently, layered lithium nickel manganese oxides, LiNi<sub>x</sub>Mn<sub>1-x</sub>O<sub>2</sub> crystallizing in the O<sub>3</sub> structure same as LiCoO<sub>2</sub> (space group R<sub>m</sub>) have been extensively investigated and can be considered as the alternative material for the commercialized LiCoO<sub>2</sub> [3-6]. However, the work regarding LiNi<sub>x</sub>Mn<sub>1-x</sub>O<sub>2</sub> is predominantly concerned with the Mn-rich type which normally shows relatively low discharge capacity and voltage.

Another type of compound with excess Li, Li-rich layered cathodes formulated as Li<sub>1+x</sub>M<sub>1-x</sub>O<sub>2</sub> (M = Mn, Ni, and/or Co) receive increasing attention due to high specific capacity (~280 mAh/g) and energy density (~1000 Wh/kg) [7-11]. Most of them are focused on the cathodes with high manganese contents which however show relatively low discharge voltages compared to the conventional LiCoO<sub>2</sub> [12]. Furthermore, Mn-rich Li<sub>1+x</sub>M<sub>1-x</sub>O<sub>2</sub> compounds are normally faced with serious voltage fade issue, making it very hard to be commercialized. Recently, it is found that increasing the Ni content in Li<sub>1+x</sub>M<sub>1-x</sub>O<sub>2</sub> could suppress discharge voltage fade and simultaneously enhance the cyclic performance significantly [13]. The increase of Ni content can increase both the capacity and voltage but may enhance the occupation of Ni at the Li lattice site and thus lead to the sluggish kinetics of Li<sup>+</sup> transport [14]. Fortunately, the excess Li is found to suppress the occupation of Ni at the Li site effectively which is expected to favor the performance of Li<sub>1+x</sub>M<sub>1-x</sub>O<sub>2</sub> [14,15].

Therefore, the incorporation of excess Li and high Ni is anticipated to probably make Li<sub>1+x</sub>M<sub>1-x</sub>O<sub>2</sub> perform well. Although little is known about the high Nickel Li-rich Li<sub>1+x</sub>M<sub>1-x</sub>O<sub>2</sub> some groups have successfully synthesized the single phase high Nickel Li-rich Li<sub>1+x</sub>M<sub>1-x</sub>O<sub>2</sub> [14,16,17]. Li et al. reported on the successful synthesis of Li<sub>1+x</sub>(Ni<sub>y</sub>Mn<sub>1-y</sub>)<sub>1-x</sub>O<sub>2</sub> (y=0.2, 0.33, 0.4, 0.5 0.6, and 0.7, 0 ≤ x ≤ 0.34) with the discharge capacity of about 200 mAh/g [16]. The Ti additive is found to be capable of enhancing the cyclic performance of Li<sub>1.167</sub>Ni<sub>0.4</sub>Mn<sub>0.383</sub>Co<sub>0.05</sub>O<sub>2</sub> effectively [17]. Again, the lithium-rich high nickel core shell structures comprised of nanoparticles were reported to have the discharge capacities around 220 mAh/g and the fairly good cycle performances [14]. However, despite the high capacity and high discharge voltage the lithium-rich high nickel layered cathode tends to

cause the decomposition of electrolyte due to the possible catalyzing effect of nickel ions with high content.

To sum up, the high Nickel-Lithium rich layered cathode has the high capacity and high discharge voltage which is very promising as candidates for next generation LIBs although it may lead to the possible decomposition of electrolyte. However, its performance is believed to be improved by alien element doping, nanostructure design and surface coating as already done for other cathode materials.

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National Center for Nano science and Nanotechnology of China, China

Correspondence: Weiguo Chu, National Center for Nanoscience and Nanotechnology of China, China, E-mail: wgchu@nanoctr.cn

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