



## Nickel-rich electrodes for Li-ion Batteries

Group 3

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### Abstract

Ni-rich electrodes are gaining increased attention in current research due to their lower cost and higher capacity. This work provides a comprehensive overview of Ni-rich electrodes for lithium-ion batteries (LIB), focusing on the structural configurations, degradation mechanisms, and commercial applications of Ni-rich materials. Many of the Ni-rich positive electrodes have performance limitations due to degradation mechanisms during cycling, and finding a suitable electrolyte is a challenge for a less degradable alternative. There is also the need of replacing cobalt in already commercialised electrodes. All these issues pose challenges to commercial application of the new Ni-rich electrode materials.

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Keywords: LIB, Ni-rich electrode, NMC, NCA, LMNO

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## 1 Introduction

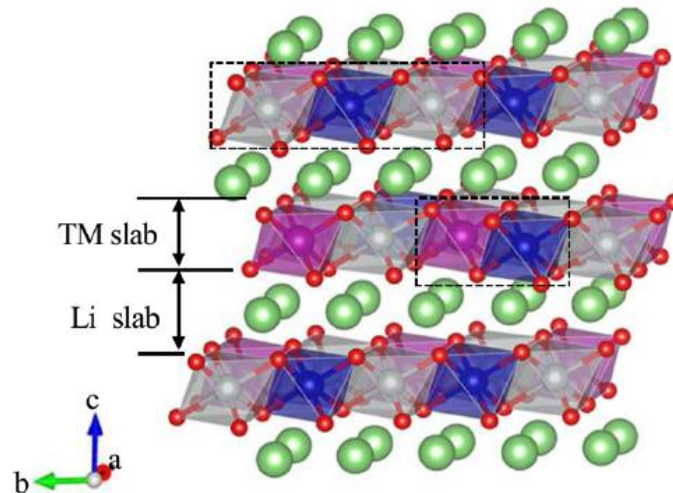
Since the 1970s, intercalation chemistry has gained increasing interest, motivated by Whittingham's discoveries leading to the first rechargeable lithium battery utilizing a titanium disulfide positive electrode ( $\text{LiTiS}_2$ ) and a lithium metal negative electrode [1]. This configuration faced limitations, such as a cell voltage limited to  $< 2.5$  V, restricting energy density, and the potential for dendrite growth on lithium-metal anodes, posing a fire hazard [2]. Goodenough's group made a significant leap in the 1980s, introducing three classes of oxide cathodes [3]–[5], elevating the operating voltage to  $\sim 4$  V and eliminating the need for metallic lithium in the negative electrode [2]. In 1991, Sony Electronics achieved the first commercial LIB with  $\text{LiCoO}_2$  as the cathode and graphite as the anode [6]. However, cobalt's limited availability and high market price lead to the exploration of alternative positive electrode materials. While  $\text{LiNiO}_2$  offers high capacity and energy density, it exhibits poorer performance and thermal stability [7]. Manganese, while cost-effective with a spinel structure, suffers from  $\text{Mn}^{3+}$  disproportionation to  $\text{Mn}^{4+}$  and  $\text{Mn}^{2+}$ , leading to cathode degradation and graphite anode poisoning, significantly limiting cell cycle life [2].

To meet the above challenges, researchers subsequently developed a series of layered oxide materials such as  $\text{Li}[\text{Ni}_{1-x-y}\text{Co}_x\text{Mn}_y]\text{O}_2$  (NMC) [8] and  $\text{Li}[\text{Ni}_{1-x-y}\text{Co}_x\text{Al}_y]\text{O}_2$  (NCA) [9]. Leading to lower cost and superior specific capacity. While the theoretical capacity of these layered cathode materials is as high as  $275 \text{ mAh g}_{\text{NMC}}^{-1}$ , not all of it is achievable due to structural instabilities occurring when an exceedingly large fraction of lithium is removed. In practice, the charging cut-off is limited to  $\sim 80\%$ – $90\%$  state of charge (SoC) depending on the application to limit positive electrode degradation [10]. To enhance accessible capacity at reasonable upper cut-off voltages, Ni-rich electrodes ( $>80\%$ ) have recently gained significant interest. More lithium can be extracted from their structure within the same voltage window while penalizing thermal stability and capacity retention [11].

Today, various positive electrodes, including Ni-rich electrodes like NMC and NCA, as well as the emerging LMNO, are available for Li-ion batteries. Over the last decade, the Ni-based layered oxides solidified their status as the cathode material of choice, especially in the electrical transport sector [12]. A smaller share of the market is instead claimed by olivine  $\text{LiFePO}_4$  (LFP) [13]. Characterized by lower energy densities, it is a stable and cheap material who is limited by low ionic and electric conductivity that can be improved by synthesis methods [14]. This however will not be furtherly characterized as not part of the Ni rich materials family. Finally high voltage (up to 5V) ordered spinel like  $\text{LiNi}_0.5\text{Mn}_{1.5}\text{O}_4$  (LNMO) [8], [15], [16] is a promising candidate as the positive electrode in next generation LIB but further developments in terms of electrolyte stability and capacity fading are required [17]. This study focuses on these electrodes, categorized into spinel and layered structures, providing insights into their characteristics, potential applications, and current limitations.

## 2 Structural changes in layered and spinel materials

### 2.1 Layered structures



*Figure 1. TM–O octahedral complexes in NMC. Ni, Mn, and Co reside at the octahedral site surrounded by six O atoms. The grey, blue, and purple spheres represent Ni, Co, and Mn atoms in the TM layer, respectively, and the red sphere represents O atoms [18].*

The class of NMC materials presents the same  $\alpha$ - $\text{NaFeO}_2$  (R3m) hexagonal structure of  $\text{LiCoO}_2$  and  $\text{LiNiO}_2$  where Li, O, and transition metals (TMs) occupy alternating atomic layers [19]. The presence of Ni, Mn, and Co contributes to the enhanced performance of NMC with Ni allowing for high discharge capacity, Mn maintaining better cycle life and thermal safety while Co providing better rate capability [18]. Still the presence of Ni and Mn is associated with some challenges related to their already discussed pure configurations.

The skyrocketing prices and increased demand for TMs, especially Co, have caused a shift towards materials richer in Ni composition, even though Ni is widely viewed as the root cause of instabilities and capacity degradation [7]. This is clearly displayed in the figure below (see Figure 2) where materials with increasing shares of Ni show higher capacities but poorer operational performances.

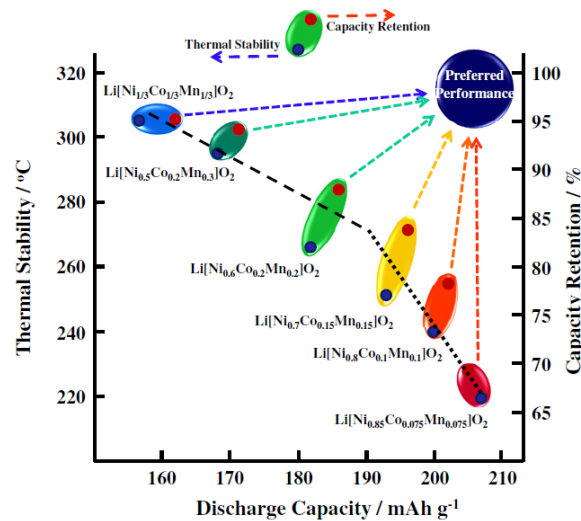


Figure 2. A map of relationship between discharge capacity, and thermal stability and capacity retention of  $\text{Li/Li}[\text{Ni}_x\text{Co}_y\text{Mn}_z]\text{O}_2$  ( $x = 1/3, 0.5, 0.6, 0.7, 0.8$  and  $0.85$ ) [11].

Just like NMC, NCA presents a layered R3m structure. This comes with both its positive and negative implications as this is a material with good cyclability and capacity despite the low Co content but keeps the structural instabilities issues [20].

## 2.2 Spinel and comparison to layered structures

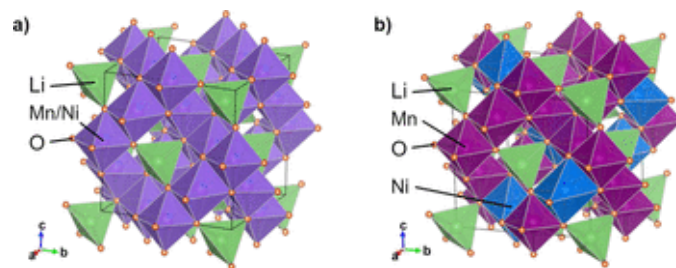


Figure 3. a) disordered LMNO, b) ordered LMNO. [21]

Because of the previously mentioned issues with thermal stability of  $\text{LiNiO}_2$ , it is not a suitable electrode material. Introducing other ions into the structure makes lithium nickel oxides easier to synthesize, more stable upon lithiation and increases the capacity, such as NMC and NCA [22]–[24]. However, due to the cobalt content in these materials groups another promising electrode material has gained more attention.  $\text{LiMn}_{1.5}\text{Ni}_{0.5}\text{O}_4$  also referred to as LMNO, is a cobalt-free positive electrode material known for its high voltage range and stable cyclability [25]. However, the many and varying crystal structures of LMNO have made the area more complex to research and even harder to commercialize. LMNO is not a layered structure such as NMC and NCA but a spinel structure. There are also multiple types of LMNO spinel structures which are divided into two forms; disordered and ordered [23].

During lithiation and delithiation, all electrode materials undergo phase transitions. As the chemical structure of LMNO is quite different from that of NMC these phase transitions are much different. For layered structures the material changes through a series of two-phase reactions according to  $H1 \rightarrow M \rightarrow H2 \rightarrow H3$ , where M is a monoclinic structure while the other phases are hexagonal crystal, seen in Figure 4 [24].

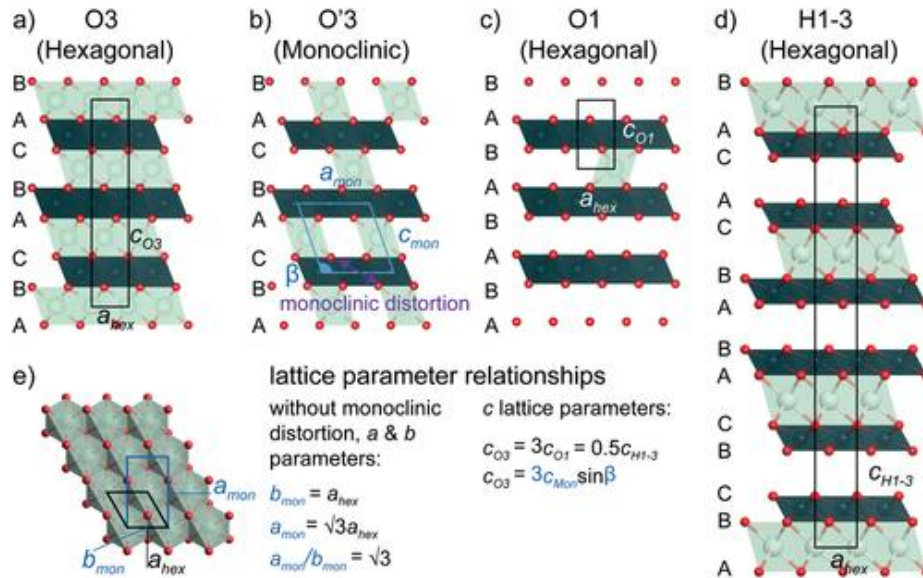


Figure 4. a) H1, b) monoclinic, c) H2, d) hybrid of H1 and H2 referred to as H3.

The complex nature of the spinel structure results in multiple crystal transformations during cycling. In broad terms, these transformations result in three major changes, cationic ordering for some specific sites, anionic ordering for other specified sites and lastly displacement of all atoms.

There is also an observed difference in electrochemical behavior between ordered and disordered spinel structures of LMNO. The disordered version gives a rise to two voltage plateaus during cycling, while the ordered one only displays one voltage plateau. Even though all the three above-mentioned changes apply to both the disordered and ordered versions, the species are chemically different. For example, the disordered version has a higher number of defects [23], [24].

However, it is the spinel structure that provides many of LMNOs unique properties. The 3-dimensional porous structure allows for fast movement of the lithium ions, which also makes it a strong candidate for fast-charging batteries. Therefore, LMNO is an up-and-coming material for high voltage applications even though it is not commercial yet [23]. The main challenge for this electrode material is the instability of the electrolyte at such potentials, which originates decomposition products that accelerate the aging of the cell [26].

### 3 Degradation mechanisms

Despite offering high specific capacity, Ni-rich layered oxide electrodes are afflicted by numerous performance degradation processes which are listed and discussed below.

#### 3.1 Cation mixing

Cation mixing is a process where metal ions with a similar ionic radius to that of  $\text{Li}^+$  occupy the sites of  $\text{Li}^+$  in the lithium layer. In the case of Ni-rich electrodes,  $\text{Ni}^{2+}$  ions have the strongest tendency to mix with  $\text{Li}^+$  ions. During the first charge, the positive electrode can be fully delithiated, however, upon discharge, part of the  $\text{Li}^+$  cannot return to the positive electrode due to the Li/Ni mixing which causes a decrease in capacity and the  $\text{Li}^+$  mobility is reduced. The cation mixing occurs both during the synthesis process and during the cycling process, and its intensity increases with the Ni content, state of charge and temperature. Due to this process, the crystal structure also changes from layered phase to spinel-like phase and finally to rock-salt phase. The formed rock-salt phase has poor transportation kinetics, which results in the blocking of  $\text{Li}^+$  diffusion paths and in the reduction of ionic conductivity [27]–[29].

#### 3.2 Residual lithium compounds

Residual lithium compounds in the form of  $\text{Li}_2\text{O}$ ,  $\text{LiOH}$  and  $\text{Li}_2\text{CO}_3$  are found in Ni-rich electrodes. These are generated either during the synthesis process or during storage due to reactions with  $\text{H}_2\text{O}$  and  $\text{CO}_2$  in the air. Besides consuming lithium, these compounds are responsible for gelation during slurry preparation and for battery swelling [30].

#### 3.3 Oxygen evolution reaction

As previously mentioned, during cycling all Ni-rich layered oxide electrodes undergo several phase transitions. The H3 phase is a thermodynamically unstable phase and is usually not reached unless the content of Ni is particularly high: the potential for the  $\text{H}_2 \rightarrow \text{H}_3$  transition decreases with an increase in the Ni content. It is observed that oxygen and carbon dioxide evolutions initiate when this transition starts [31]. The oxygen evolution reaction is believed to be one of the main causes for depletion of electrolyte solvents and cathode structural deterioration. Similarly, to what happens due to cation mixing, with the loss of oxygen, the crystal structure of the electrode is changed from layered phase to spinel-like phase and rock-salt phase [27].

#### 3.4 Transition metal dissolution

An additional side effect of both the cation mixing and the oxygen evolution reactions is the transition metal ion dissolution, which is observed in the H3 phase. This results in the depletion of active transition metal ions, leading to a reduction in capacity. Moreover, the ions can migrate to the negative electrode side contributing to the SEI and increasing its impedance [27], [31].



### 3.5 Generation of micro cracks

Finally, because of the many changes in the structure of the electrode, microcracks along the grain boundaries are generated. These cracks, besides increasing the impedance of the electrode, also allow the liquid electrolyte to penetrate, leading to a degradation of the battery performance [27], [28].

For each of the above-discussed processes, various strategies such as doping of foreign ions, surface coating, crystal and morphology structure regulation can mitigate the degradation effects [28].

## 4 Commercial application and limitation of Li-ion batteries with Ni-rich electrodes

The degradation mechanisms of Ni-rich electrodes pose challenges to commercial application of them, especially causing capacity fade over a longer period of uses. In this section, a summary of reported limitations of commercially available LIB with Ni-rich electrodes is presented. Capacity fade of commercial NCA and NMC cells will be compared with a LFP cell, their main market competitor for positive electrode for batteries in EV applications.

### 4.1 Temperature dependency

Degradation of NMC and NCA cells takes place in different mechanisms depending on the temperature. It has been reported that the dominant aging mechanism is Li plating below 25 °C and SEI growth above 25 °C for an NMC/LMO-graphite cell. The temperature at which the degradation rate is the lowest and transition of the dominant degrading mechanism is observed, 25°C in this case, is referred to as the tipping point. For NMC cells, 35 °C is consistently reported as the tipping point [32], [33], while NCA cells do not seem to have a strong dependence on temperature in the range of 15 °C to 35 °C [34].

Cycling tests on commercial NMC and NCA cells<sup>1</sup> showed that at even their tipping points, i.e., at the optimal temperatures for their performance, both cells exhibited much faster degradation rate when compared to a LFP cell manufactured in the same format [34]. Tested NMC and NCA cells reached 80% of their initial capacity after around 600 and 400 equivalent full cycles of 20%–80% SoC, respectively, while the LFP cell maintained approximately 95% of its initial capacity even after 2000 cycles in the same test.

### 4.2 Discharge rate dependency

On the other hand, for NCA cells, increasing discharge rate resulted in slower degradation [34], [35], while degradation of LFP and NMC seemed not to be affected. However, it has not been

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<sup>1</sup> NCA cell from Panasonic, 3.2 Ah; NMC cell from LG Chem, 3 Ah. Both cells are in 18650 format.

elucidated why the NCA cells are more strongly influenced by the discharge rate than the other two.

#### **4.3 DOD dependency**

Lastly, capacity fade of a cell can be accelerated by an increasing depth of discharge, because of larger changes in the volume of the graphite during the intercalation [32]. Compared to a LFP counterpart, it was observed that commercial NCA and NMC cells are more affected by DOD, which could be an effect of their higher operating voltages than that for LFP [36].

These findings reflect the main challenges that must be overcome for LIB with Ni-rich electrodes to satisfy high energy and stability at commercial scale. Nevertheless, commercial LIB with Ni-rich electrodes are still widely deployed, especially in the automotive market, motivated by their cost competitiveness due to increasing price of cobalt and high energy capacity that enables longer drive range compared to other alternatives such as LFP (LiFePO<sub>4</sub>) batteries [12], [37]. For example, Panasonic/Tesla's NCA92 positive electrode, which was employed for batteries for Tesla's Model 3 in 2017, reduced the cost by 50% in comparison to LCO [38]. Even with increasing cost and persistent necessity of cobalt, it is believed that the market for Ni-rich electrodes will continue to grow [37], [39].

### **5 Conclusion**

The high capacity and the low cost of Ni-rich electrodes makes them a promising alternative for the next generation Li-ion batteries. However, several degradation mechanisms introduce challenges to their commercial applications. Cycling tests on commercially available LIB with NMC and NCA cells revealed that the capacity fade rate of the LIB with Ni-rich electrodes is much faster and more sensitive to temperature and SoC range compared to LFP cells, eventually limiting their lifespan. LMNO is a promising candidate that has little degradation, however, finding a suitable electrolyte that does not decompose within the voltage range remains as a challenge.



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