

# Research Progresses and Development Prospects on Cathode Materials for Lithium-Ion Batteries

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**Abstract:** Fossil fuels are in short supply, and people are trying to find new energy sources to replace them. Recently we have an increasing demand for high energy density lithium-ion batteries, especially in the field of daily life, such as electric cars and other. The existing cathode material technology has become a limiting factor in the discovery of lithium-ion batteries. Accordingly, it is of extraordinary significance to conduct in-depth research on high-performance and high-energy-density cathode materials. Cathode materials at this stage have their own characteristics. However, they are still immature in many ways like security, steadiness and inner framework. A lot of recent research revolves around the discovery of lithium-ion battery cathode materials with greater cycle life, higher energy density, and greater stability. Herein, the structure and mechanism of several main cathode materials, as well as their problems in charge and discharge are summarized, and the recent studies on improving electrochemical properties of cathode materials by modification are reviewed.

## 1 INTRODUCTION

As our modern science and technology continues to advance rapidly, the immense utilization of fossil fuels is triggering increasingly nasty climate variability and global warming (Zhao et al, 2021). Because the development of sustainable energy storage technology is crucial to building a low-carbon society, lithium-ion batteries (LIBs) have drawn rising interest (Li et al, 2022). In general, LIBs have notable benefits like high energy densities, efficient cycling performance, and rapid response times. Their suitability for energy storage ranging from kilowatts to megawatts with swift response times is acknowledged. Furthermore, LIBs automotive supply chain is global and has a mature infrastructure in place, making it highly desirable among consumers. These existing capabilities could serve as a cornerstone for the accelerated deployment of LIBs in grid-scale energy storage applications (Yang et al, 2022). Recently, we've noticed an increasingly obvious problem surge in the advancement of electric vehicles (EVs). Simultaneously, considerable research endeavors are focused on uncovering cathode materials for LIBs that exhibit continued cycle longevity, elevated energy density, and

structural stability (Cai et al, 2021).

The primary commercial cathode materials utilized in LIBs include LiCoO<sub>2</sub>, LiMn<sub>2</sub>O<sub>4</sub>, LiFePO<sub>4</sub>, and LiNiCoMnO<sub>2</sub>. Among these, LiCoO<sub>2</sub> stands out as one of the earliest materials extensively employed in portable electronics owing to its elevated operational voltage, extended cycling durability, minimal self-discharge, and eco-friendliness. Nonetheless, whether it can be used commercially or not we still need to consider its disadvantages. LiCoO<sub>2</sub> does present certain disadvantages, outlined as follows. Firstly, the limited availability of cobalt resources results in the high cost of LiCoO<sub>2</sub>. Secondly, the inherent structural instability of LiCoO<sub>2</sub> contributes to significant capacity deficiencies. Thirdly, cobalt is a toxic element, posing environmental concerns. Specifically, its discharge capacity and cycling efficiency deteriorate rapidly when operating above 4.2 volts (Chen et al, 2021). After discovering that LiCoO<sub>2</sub> could be difficult to use commercially because of its shortcomings, through years of research, researchers discovered LiMn<sub>2</sub>O<sub>4</sub>. However, the cycling performance of LiMn<sub>2</sub>O<sub>4</sub> falls short due to the Jahn-Teller effect, Mn dissolution, and electrolyte disintegration at high charge voltage (Li et al, 2021). Goodenough

discovered a commercially available cathode material that possesses a smooth voltage plateau, as evidenced by high energy density, substantial power and structural stability, and that's the  $\text{LiFePO}_4$ . Nevertheless,  $\text{LiFePO}_4$  has its drawbacks, including a relatively platforms with low charging voltage and poor conductivity. Consequently, to achieve optimal electrochemical performance,  $\text{LiFePO}_4$  requires a conductive layer coating, a process that contributes to its lower tap density.

With the discovery of anode material, many researchers created a novel material which was called layered lithium nickel cobalt manganese (NCM) that might be economically employed in LIBs (Akhilash et al, 2023). The NCM material is really classified like NCM111, NCM523, NCM622, NCM811 and so on. Each metallic element has an important purpose in the multilayer NCM, the Ni ingredient provides a high actual capacity, while the Co element adds to desirable cycle life. There is high structural stability observed in NCM, attributed to the presence of the Mn element. The electrochemical capacity of NCM shows a positive correlation with the content of Ni ions, as Ni ions are the main active redox material, and NCM has recently become one of the alternatives for commercial cathode materials.

Despite the fact that NCM exhibits several

advantages like higher reversible capacity, reduced cost, and enhanced environmental compatibility over the widely utilized  $\text{LiCoO}_2$ . It also has some notable disadvantages. For example, the durability of NCM during cycling is insufficient to satisfy the stringent demands for the extended lifespan of LIBs. Furthermore, NCM demonstrates substandard electrochemical performance under high speed charge and discharge conditions. Besides, it performs poorly thermally at high temperatures and the issue will get considerably worse as the Ni concentration rises. To address these issues, we need to continue to attack the future. In the last several years, a lot of in-depth research has been done by scientists on it to solve the problems like low energy density and short cycling life is present in the LIBS cathode materials and achieved significant advancements in performance. In conclusion, these issues may be resolved in a number of methods, including surface treatment or surface coating, ion incorporation, core-shell architecture and improved electrolyte, and more.

Hereunder, We conclude with a summary of the latest research advances on high-energy LIBs cathode materials and offer some suggestions and directions for future research. We hope that this contributes to the ongoing research efforts in the field of LIBs.

## 2 CATHODE MATERIALS

### 2.1 $\text{LiCoO}_2$

$\text{LiCoO}_2$  stands out as one of the earliest commercially available cathode materials for LIBs (Li et al, 2018). Its appeal in the market stems from its simple preparation method, high specific capacity, stable cycling performance, and good thermal stability.  $\text{LiCoO}_2$  demonstrates three primary crystal structures: layered, spinel, and rock salt phases. Of these, the ordered  $\alpha\text{-NaFeO}_2$  layered structure facilitates rapid and reversible lithium ions insertion/extraction during production expansion, making it the most utilized configuration, accounting for over 31% of lithium-ion battery usage (Qian et al, 2018).

The stratified arrangement of  $\text{LiCoO}_2$ , resembling the  $\alpha\text{-NaFeO}_2$  structure, belongs to the hexagonal crystal system, with specific lattice constants. Within this structure,  $\text{CoO}_2$  forms a two-dimensional atomic layer, with oxygen atoms adopting a cubic dense packing. Cobalt and lithium ions alternate between octahedral positions within oxygen atoms, allowing lithium ions to move between the atomic layers. However, due to varying interaction forces between

ions and oxygen atoms, the oxygen atoms deviate from an ideal cubic close-packed structure, distorting into a hexagonal close-packed structure and reducing lattice symmetry.

Despite the theoretical specific capacity is  $274 \text{ mAhg}^{-1}$ , the actual discharge capacity equals limited to  $135\sim 140 \text{ mAhg}^{-1}$ . This discrepancy arises from the charge and discharge process, involving specific reactions. During lithium ions extraction, vacancies form in the original lattice, impacting atomic arrangement and inducing a phase transition. Varying levels of lithium ions extraction generate multiple phases, with transition states marked by changes in lattice parameters. As lithium extraction increases, the crystal structure shifts from hexagonal to monoclinic, leading to local asymmetrical expansion and contraction, ultimately compromising battery cycling stability.

In practical charge and discharge processes, only approximately 53% of lithium ions can reversibly intercalate and deintercalated (Liu et al, 2020). Increasing charging voltage enhances lithium extraction, but excessive extraction damages cycling stability, necessitating a voltage limit of 4.2V (vs.  $\text{Li/Li}^+$ ). This limitation results in an actual capacity of  $135\sim 140 \text{ mAhg}^{-1}$  ( $\text{Li}_{1-x}\text{CoO}_2$ ,  $x \approx 0.5$ ,  $\sim 4.2 \text{ V}$  vs.  $\text{Li/Li}^+$ ), highlighting the need to enhance cathode

material stability under high voltage conditions (Qian et al, 2018).

## 2.2 NCM

In the realm of cathode materials for batteries, ongoing research focuses on three main structures: one-dimensional olivine, two-dimensional layered, and three-dimensional spinel. Among these, the NCM cementitious materials hold a prominent position due to their high capacity and power ratings, ideal for high-energy density battery production.

NCM shares a similar layered structure with the  $\alpha$ -NaFeO<sub>2</sub> type, residing within the hexagonal crystal system under space group R-3m. Here, nickel ions, trivalent cobalt ions, and tetravalent manganese ions occupy octahedral center positions, forming a cubic close-packed arrangement in a layered fashion (Ko et al, 2023). Ternary materials offer distinct advantages over unary and binary counterparts, thanks to the unique roles of different cations: nickel ions enhance specific capacity, trivalent cobalt ions reduce cation mixing and enhances structural stability and electronic conductivity, while tetravalent manganese ions introduce cost-effectiveness and improves thermal stability (Qian et al, 2018).

However, simply increasing cation content doesn't universally enhance material performance. For instance, heightened Co<sup>3+</sup> content may diminish NCM's reversible specific capacity and inflate battery production costs. Similarly, excessive nickel ions content risks gasification and deliquescence during charge and discharge, compromising thermal cycling stability (Yan et al, 2020). Moreover, an overabundance of tetravalent manganese ions can introduce a spinel phase, disrupting the material's layered structure and impairing rate performance (Ein-Eli et al, 1998).

Furthermore, NCM's morphology significantly influences battery volumetric energy density. While traditional lithium cobalt oxide boasts a theoretical density of 3.9 g/cm<sup>3</sup>, NCM materials hover around 3.6 g/cm<sup>3</sup> (Wang et al, 2020). To elevate volumetric energy density and voltage platform, tailored morphological modifications are essential to align with user demands.

## 2.3 LiFePO<sub>4</sub>

LiFePO<sub>4</sub> boasts a typical olivine structure characterized by robust covalent bonds between phosphorus and oxygen, imparting exceptional strength, high-temperature resistance, and chemical stability. Its three-dimensional olivine structure

fosters a one-dimensional channel for lithium ions transport, effectively constraining ion diffusion. In contrast to ternary lithium batteries, LiFePO<sub>4</sub> batteries exhibit superior structural stability and enhanced safety.

The operational principle of LiFePO<sub>4</sub> batteries hinges on the relocation of lithium ions across the electrodes of a cell to facilitate charging and discharging (Su, 2022). During charging, lithium ions disintercalate from the anode material (LiFePO<sub>4</sub>) and intercalate into the cathode material (typically graphite) via the electrolyte, enabling reversible multi-cycle charge-discharge capability. The charging process involves the positive electrode compound LiFePO<sub>4</sub>, where lithium ions disintercalate to form FePO<sub>4</sub>, liberating electrons. The negative electrode, graphite, demonstrates efficient lithium ions insertion and extraction, with lithium ions migrating into the graphite from the electrolyte while concurrently releasing electrons. Organic solvents containing lithium salts, like LiPF<sub>6</sub>, function as electrolytes, facilitating lithium ion transport (Apachitei et al, 2023).

LiFePO<sub>4</sub> batteries offer higher energy density, translating to extended range. Compared to other lithium-ion variants, they exhibit prolonged lifespan and increased charge-discharge cycle endurance. Additionally, they demonstrate robust stability, minimizing risks of overheating, combustion, and related safety concerns. Environmentally, LiFePO<sub>4</sub> batteries are advantageous as they lack heavy metals and harmful substances. However, they exhibit relatively slower charging rates and necessitate longer charging durations (Su, 2022). The relatively intricate manufacturing processes contribute to higher prices.

## 2.4 LiMn<sub>2</sub>O<sub>4</sub>

The crystal structure of lithium manganese oxide comprises monoclinic and orthorhombic phases. The monoclinic phase, akin to LiCoO<sub>2</sub> but with lower symmetry, contrasts with the more stable orthorhombic phase. Despite its inherent stability, lithium manganese oxide is susceptible to Jahn-Teller distortion, rendering it thermodynamically unstable. Cycling induces a transformation to the more stable spinel structure LiMn<sub>2</sub>O<sub>4</sub>, causing a notable reduction in reversible capacity.

In the charge-discharge cycle of lithium manganese oxide batteries, manganese ions undergo oxidation-reduction reactions. Charging oxidizes Mn<sup>+</sup> to Mn<sup>4+</sup>, liberating lithium ions, while discharging reduces Mn<sup>+</sup> to Mn<sup>3+</sup>, re-embedding

lithium ions into the lattice. This redox reaction is pivotal to the battery's operation. Lithium ions shuttle between electrodes, embedding into and extracting from the crystal structure. This process forms the core of the battery's charge-discharge mechanism (Li et al, 2021).

The spinel structure of lithium manganese oxide exhibits robust stability, enduring the embedding and extraction of lithium ions, thus contributing to cycling stability and longevity. Abundant resources, low production costs, and mature manufacturing processes make lithium manganese oxide suitable for practical applications. It offers higher specific capacity and energy density, alongside good safety and environmental attributes. However, its cycling stability is compromised by structural changes and dissolution, curtailing battery lifespan (Li et al, 2021). Jahn-Teller distortion, particle aggregation, and surface reactions further impact its electrochemical performance.

## 2.5 LiNiO<sub>2</sub>

Lithium nickel oxide adopts a layered crystal structure, featuring both rock salt and  $\alpha$ -NaFeO<sub>2</sub> configurations. Within this structure, nickel ions and lithium ions coexist. The integration of nickel ions into the lithium layers introduces additional complexity to the crystal lattice. Moreover, the phenomenon of non-stoichiometry arises due to high-temperature calcination, resulting in lithium loss from the crystal structure and deviating from strict stoichiometry.

Irreversible phase transitions occur in lithium nickel oxide, leading to the formation of an inactive NiO<sub>2</sub> phase upon charging to higher voltages, which in turn contributes to capacity decay. The distinctive characteristics of lithium nickel oxide, namely its layered structure and non-stoichiometric nature, significantly influence its electrochemical performance and cycling lifespan (Kalyani & Kalaiselvi, 2005).

During charge and discharge, trivalent nickel ions undergo oxidation to tetravalent nickel ions. However, migration of some nickel ions to lithium sites within the structure compromises cycling lifespan. Lithium nickel oxide exists in two structural forms: cubic and layered hexagonal phases, with only the latter exhibiting electrochemical activity (Kalyani & Kalaiselvi, 2005). Cycling induces phase transitions, such as the conversion from hexagonal to monoclinic phases, altering the crystal structure and impacting electrochemical performance.

The conductivity of lithium nickel oxide is closely

tied to the positioning of nickel ions within the crystal lattice. Migration of nickel ions disrupts electron conduction paths between nickel ions, reducing conductivity. Additionally, changes in nickel ion positions affect ion diffusion rates, further influencing charge and discharge performance.

With a maximum specific energy density of 160 mA/g, LiNiO<sub>2</sub> surpasses other cathode materials (Kalyani & Kalaiselvi, 2005). It demonstrates minimal capacity decay during cycling, boasting an excellent cycling lifespan. LiNiO<sub>2</sub> can withstand deep discharge conditions below 2 V, maximizing energy storage capacity. However, its phase transition process is only partially reversible and slower compared to LiCoO<sub>2</sub>, necessitating optimization of synthesis conditions to mitigate these challenges.

## 3 MODIFICATION METHODS FOR CATHODE ELECTRODE

Current research on cathode electrode materials primarily focuses on doping modification and surface coating of high-nickel ternary and lithium-rich cathode electrode materials, as well as their structural evolution during extended cycling.

### 3.1 Surface Modification

Many experts are looking for the ways of surface coating to improve performance. For the modification of surface coatings nowadays, in order to enhance their corrosion resistance, electrical conductivity, abrasion resistance and structural optimisation, the main methods are the selection of coating materials with excellent properties, such as polymers, ceramics and metal oxides. Or to carry out coating design to design coatings with optimised structure and thickness. Or proper cleaning, roughening or functionalisation of the cell surface before applying the coating to enhance the adhesion of the coating. We have also summarised some studies as follows. For example, Tan et al. utilized ZrTiO (ZTO) as an epitaxial layer to enhance the mechanical stability of ultra-high nickel LiNi<sub>0.8</sub>Co<sub>0.1</sub>Mn<sub>0.1</sub>O<sub>2</sub> (NCM90). Structural characterization and multi-field analysis revealed that the congruent ZTO layer and Zr doping effectively suppressed internal strain and lattice oxygen release, inhibiting local stress accumulation during cycling. The protective ZTO layer also prevented electrolyte erosion, preserving the intact surface structure of NCM90. NCM90 exhibited improved cycling Voltage endurance (4.5 V),



possessing a capacity retention rate of 71% after 100 cycles. Also, Wu et al. proposed a powder coating method, exemplified by coating a nickel cathode with  $\text{Al}(\text{OH})_3$  nanoparticles, to illustrate the chemical changes on the cathode surface with varying coating levels. This coated cathode exhibited improved cycling stability and rate capability, attributed to effective surface protection against side reactions and enhanced lithium ions transport. Furthermore, environmental and thermal stability were enhanced.

### 3.2 Doping Modification

There are also experts who work on improving material properties through doping modification. Zhang et al. addressed irreversible phase transitions and cracking issues in  $\text{LiNiO}_2$ -based cathode electrodes by doping single crystal  $\text{LiNi}_{0.83}\text{Co}_{0.07}\text{Mn}_{0.1}\text{O}_2$  (SNCM) with tungsten boride (WB). Through microscopy and spectroscopic evidence, they demonstrated that Trivalent boron ions entering the SNCM matrix expanded the interlayer spacing, facilitating lithium ions diffusion, while  $\text{W}^{3+}$  formed a non-crystalline surface layer composed of  $\text{Li}_x\text{W}_y\text{O}_z$  (LWO) and  $\text{Li}_x\text{B}_y\text{O}_z$  (LBO), contributing to the creation of a robust cathode electrolyte interphase (CEI) membrane structure. WB doping effectively controlled c-axis contraction and oxygen gas release at high voltages, with an optimal doping concentration of 0.6 wt.% resulting in 93.2% capacity endurance after 200 cycles at 2.7–4.3 V. Also, Fan et al. introduced a method to substitute Transition-element cations in  $\text{LiNi}_{0.83}\text{Co}_{0.11}\text{Mn}_{0.06}\text{O}_2$  with Mo-NCM, investigating the preparation structure evolution due to Mo substitute and the constancy of the Mo-NCM positive electrode. Experimental and computational results revealed that partial substitution with hexavalent molybdenum ions induced cation ordering and facilitated lithium ions diffusion kinetics, with pentavalent molybdenum ions cations acting as pillars to avoid local failure and structural distortion. Strong Mo-O bonds stabilized lattice oxygen, enhancing the chemical durability of the nickel-rich positive electrode. Besides, Huang et al. proposed a TSS together with the addition of crystallization modifiers, including phase generation, grain enlargement, and phase retention.  $\text{MoO}_3$  addition inhibited grain enlargement and cluster formation, leading to highly dispersed Mo surface-doped single crystal NCM cubes (MSC-NCM) with an mean particle count diameter of 1.3  $\mu\text{m}$ . Lithium ions migration preferred the (104) crystal plane in MSC-NCM cubes due to surface molybdenum doping and reduced surface energy, resulting in

significantly improved rate performance compared to conventional single crystal NCM materials (SC-NCM). Furthermore, Li et al. introduced a method involving ammonium niobium oxalate treatment to create oxygen vacancies and pentavalent niobium ions doping on cobalt-free lithium-rich layered oxide (LRO) materials. The modified sample, LRO-Nb-0.5, exhibited exceptional electrochemical capability, providing a discharge capacity per unit mass of 209.77 mAh/g at 1C, with a capacity retention rate of 93.22% after 300 cycles. Moreover, LRO-Nb-0.5 demonstrated enhanced rate performance, achieving a discharge specific capacity of 178.70 mAh/g at 5 C. Pentavalent niobium ions doping and oxygen vacancy regulation emerged as crucial approaches to enhancing performance metrics of cobalt-free lithium-rich cathode materials in electrochemical applications.

### 3.3 Structure Modification

There are many experts who have changed the structure to achieve this in order to improve its performance, and the results show that it is indeed a feasible method. For example, Tan et al. integrated a tenon-mortise structure into the layered phase of  $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$  (NCM811), mitigating volume changes and facilitating lithium ions transport. The discharge capacity reached 215 mAh/g at 0.1 C with an initial Coulombic efficiency of 97.5%. After 1200 cycles at 1 C, the retention of capacity rate was 82.2%. Also Shi et al. investigated the degradation process of single-crystal high-nickel cathode material (SCNCM) during room temperature storage. Exposure to air led to a changing from a layered to a NiO rock salt phase, resulting in decreased electrochemical performance due to oxygen precipitation and the formation of oxygen vacancies. Besides, Zhao et al. studied quasi-single-crystal  $\text{LiNi}_{0.65}\text{Co}_{0.15}\text{Mn}_{0.20}\text{O}_2$  (SC-NCM65) and found that maintaining a cutoff voltage of 4.6 V preserved over 77% capacity retention after 400 cycles, while at 4.7 V, capacity dropped to 56%. SC-NCM65 degradation was primarily due to rock salt phase accumulation on particle surfaces, increasing impedance and transition metal dissolution. The thickness of the rock salt layer exhibited a linear relationship with capacity loss, indicating charge transfer kinetics as a crucial factor hindered by lower lithium ions diffusion in the NiO phase. Additionally, Li et al. introduced a method involving ammonium niobium oxalate treatment to create oxygen vacancies and pentavalent niobium ions doping on cobalt-free lithium-rich layered oxide (LRO) materials. The modified sample, LRO-Nb-0.5, exhibited exceptional

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## 4 CONCLUSION AND OUTLOOK

Among these batteries, LIBs have relatively high energy density and high charge/discharge efficiency, but they cannot withstand high temperatures. The lithium iron phosphate battery can make the cathode structure more stable, which makes its cycle life longer than other kinds of batteries, and has a lower cost. However, the energy density and charge/discharge efficiency of lithium iron phosphate batteries are low, and their performance is poor at low temperatures. In order to improve the shortcomings of these batteries, we have also searched and summarised some modification methods such as surface modification treatment, ionic doping, improved electrolyte and so on. However, the existing methods can't solve all the battery problems well, and we still need to carry out in-depth research on safety issues and internal structure of batteries in the future.

And for the existing problems, we summarise some directions for research to solve them. Regardless of whether the energy density of NCM has been developed to the upper limit or not, it is obvious that it cannot meet the further development needs of battery energy storage, so we need to find ways to develop new cathode materials with high Ni content. In this process, how to replace the Co element, how to control the cost, and how to save the limited resources of the earth are also huge challenges for us. In addition, for some of the modification methods we are now researching and developing, although there is some improvement in electrochemical performance, the cost is difficult to control, and the industrial process is difficult to achieve commercially. Therefore, we can put more effort into new types of batteries, such as lithium-sulfur batteries and solid-state batteries, compared with traditional lithium-ion batteries. In conclusion, a simple and feasible modification scheme is crucial in developing high energy density batteries in the future. However, I believe that with the development of technology and

further research on batteries, we will eventually solve these problems well.

## AUTHORS CONTRIBUTION

All the authors contributed equally and their names were listed in alphabetical order.

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