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Literature Review: Strategies for Enhancing the Performance of Li $[Ni_xCo_yMn_{1-x-y}]$ O_2 Cathode Materials for Li-Ion Batteries

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Abstract

$Li[Ni_xCo_yMn_{1-x-y}]O_2$ layered oxides Cathode Materials are among the most widely studied cathode materials for lithium-ion batteries due to their high gravimetric and volumetric energy density compared to other type cathode materials. However, the practical deployment of Ni-rich NCM materials is hindered by severe degradation mechanisms, including cation-mixing, surface reconstruction, electrolyte reactivity, transition metal dissolution, and oxygen release, which compromise cycling stability and safety. This review systematically synthesizes recent progress in advanced modification strategies designed to mitigate degradation in $Li[Ni_xCo_yMn_{1-x-y}]O_2$ cathodes. The discussion is structured into four major approaches: (i) surface modification, which employs protective coatings to suppress interfacial reactions and stabilize the cathode–electrolyte interphase; (ii) elemental doping, which strengthens the lattice, reduces cation mixing, and inhibits oxygen evolution; (iii) single-crystal engineering, which eliminates grain-boundary failure and improves thermal stability; and (iv) concentration-gradient architectures, which alleviate internal stress and enhance the durability of Ni-rich cathodes. Empirical evidence demonstrates that these strategies not only extend cycle life but also provide mechanistic insights into the underlying degradation pathways. By consolidating findings from recent experimental, this review highlights the necessity of integrating structural, chemical, and morphological interventions to realize the full potential of Ni-rich NCM cathodes. The insights presented offer a framework for designing safer, higher-performance, and commercially scalable lithium-ion batteries.

Keywords: $Li[Ni_xCo_yMn_{1-x-y}]O_2$, Lithium-ion Battery, Surface Coating, Elemental Doping, Single-Crystal Structure, Concentration Gradient, Degradation Mechanisms

Abstrak

$Li[Ni_xCo_yMn_{1-x-y}]O_2$ berbasis oksida berlapis merupakan salah satu material katoda yang paling banyak dikaji dalam pengembangan baterai litium-ion. Keunggulan utamanya terletak pada kerapatan energi gravimetri dan volumetri yang lebih tinggi dibandingkan dengan jenis katoda lain. Namun demikian, penerapan praktis material NCM kaya nikel masih menghadapi sejumlah kendala serius akibat berbagai mekanisme degradasi, antara lain pencampuran kation, rekonstruksi permukaan, reaktivitas dengan elektrolit, pelarutan logam transisi, serta pelepasan oksigen. Mekanisme-mekanisme tersebut secara langsung menurunkan stabilitas siklus dan tingkat keselamatan baterai.Ulasan ini menyajikan sintesis sistematis mengenai perkembangan mutakhir strategi modifikasi lanjutan yang dirancang untuk menekan degradasi pada katoda $Li[Ni_xCo_yMn_{1-x-y}]O_2$. Terdapat empat pendekatan utama yang dibahas, yaitu: (i) modifikasi permukaan, melalui penerapan lapisan pelindung guna menekan reaksi antarmuka dan menstabilkan lapisan katoda–elektrolit; (ii) doping unsur, yang berfungsi memperkuat struktur kisi, mengurangi pencampuran kation, serta menekan evolusi oksigen; (iii) rekayasa kristal tunggal, yang mengatasi kegagalan pada batas butir sekaligus meningkatkan stabilitas termal; dan (iv) arsitektur gradien konsentrasi, yang mampu meredam tegangan internal dan memperpanjang daya tahan katoda kaya nikel. Hasil-hasil empiris menunjukkan bahwa penerapan strategi tersebut tidak hanya memperpanjang umur pakai siklus, tetapi juga memperkaya pemahaman tentang mekanisme degradasi yang mendasari. Dengan mengintegrasikan temuan-temuan eksperimental terbaru, ulasan ini menegaskan pentingnya perpaduan intervensi struktural, kimia, dan morfologis untuk mengoptimalkan kinerja katoda NCM kaya nikel. Wawasan yang dihadirkan sekaligus menawarkan kerangka konseptual bagi pengembangan baterai litium-ion yang lebih aman, berkapasitas tinggi, dan memiliki prospek komersialisasi yang luas.

Kata kunci: $Li[Ni_xCo_yMn_{1-x-y}]O_2$, Baterai Litium-ion, Pelapisan Permukaan, Doping Unsur, Struktur Kristal Tunggal, Gradien Konsentrasi, Mekanisme Degradasi

1. Introduction

The rapid expansion of global energy demand has long been sustained by fossil fuels such as coal, oil, and natural gas (Byeon et al. 2024). While renewable energy technologies continue to grow, fossil energy sources still dominate the global energy portfolio, raising concerns about finite reserves, carbon emissions, and environmental degradation. As nations pursue sustainable energy transitions, electrification of transportation has emerged as a cornerstone strategy to mitigate dependence on fossil fuels and reduce greenhouse gas emissions (Kaushik et al. 2024; Wang et al. 2023; Xu et al. 2020). Among these efforts, electric vehicles (EVs) have gained remarkable traction, with sales surging from thousands in the early 2010s to millions annually by the 2020s. This accelerating adoption has placed lithium-ion batteries (LIBs) at the center of technological innovation, as they remain the most viable energy storage solution for EVs due to their high efficiency, long cycle life, and scalability (Byeon et al. 2024; Morchhale et al. 2023).

Despite their advantages, LIBs face limitations in energy density and cost, primarily dictated by the properties of cathode materials, which contribute nearly 40% of the total battery cost. Conventional cathode chemistries such as $LiCoO_2$, $LiFePO_4$, and $LiMnO_4$ have played significant roles in consumer electronics and early EVs, but their modest specific capacities and limited energy densities restrict their potential for next-generation applications. $LiCoO_2$ offers good electrochemical stability but suffers from high cost, resource scarcity, and safety concerns. $LiFePO_4$ provides outstanding stability and long cycle life but is hindered by low conductivity and limited volumetric energy density. $LiMn_2O_4$ delivers excellent ionic transport and structural stability, yet its capacity degradation during long-term cycling undermines its viability in high-energy applications (Byeon et al. 2024).

To overcome these constraints, layered oxide cathodes especially nickel-rich lithium nickel–cobalt–manganese oxides ($Li[Ni_xCo_yMn_{1-x-y}]O_2$, NCM) have emerged as the most promising candidates. Ni-rich NCM materials (e.g., NCM811, NCM622, and their derivatives) offer higher reversible capacities (>200 mAh g^{-1}) and specific energies approaching 800 Wh kg^{-1}, outperforming conventional cathodes. They also provide flexibility in reducing cobalt content, addressing both cost and ethical concerns related to cobalt mining. Notably, Ni-rich compositions, such as those employed in commercial EVs (e.g., Tesla's NCA/NCM chemistries), have demonstrated extended driving ranges and competitive performance metrics (Xu et al. 2024).

However, the high nickel content that enables superior capacity also introduces significant challenges. Ni-rich NCM cathodes are prone to structural instabilities, surface degradation, oxygen release, transition metal dissolution, cation mixing, and particle cracking during cycling. These degradation pathways accelerate capacity fade, reduce safety, and limit commercial scalability. Consequently, developing advanced strategies to stabilize Ni-rich NCM materials has become a focal point of contemporary battery research (Zheng et al. 2023).

In recent years, four primary approaches have gained prominence (Byeon et al. 2024; Li et al. 2024): (i) surface modification, including protective coatings to suppress parasitic reactions; (ii) elemental doping, to reinforce lattice stability and reduce cation disorder; (iii) single-crystal engineering, which mitigates grain boundary failure and improves structural integrity; and (iv) concentration-gradient architectures, which balance mechanical stress and enhance long-term cycling performance. Collectively, these strategies not only extend the practical lifetime of Ni-rich cathodes but also provide critical insights into the fundamental mechanisms of degradation.

This review consolidates and evaluates recent progress in these advanced modification strategies, with emphasis on their mechanisms, advantages, and limitations. By integrating evidence from experimental, the discussion aims to provide a comprehensive framework for understanding the pathways to stabilize Ni-rich NCM cathodes and to guide the design of next-generation high-energy lithium-ion batteries.

2.Methodology

2.1. Data Sources

The primary data for this review were collected from reputable scientific databases, including Scopus, Web of Science, ScienceDirect, SpringerLink, and Google Scholar as a supplementary source. Only peer-reviewed journal articles, experimental reports, and prior reviews directly related to strategies for improving Nickel–Cobalt–Manganese (NCM) cathode materials in lithium-ion batteries were considered.

2.2. Analytical Approach

A narrative and thematic analysis framework was employed to systematically organize and evaluate the selected literature. The review process involved:

1. Identifying core themes in the existing body of work, particularly focusing on (a) surface modification and coating, (b) doping strategies, (c) single-crystal strategies, and (d) concentration-gradient structures.
2. Categorizing the studies based on structural modification approaches and electrochemical performance (capacity, cycle retention, rate capability, and thermal stability).
3. Comparative assessment of findings across different studies to highlight converging evidence, contrasting outcomes, and emerging research directions.

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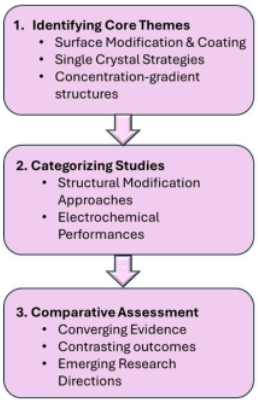

Fig. 1 Flowchart of the Narrative and Thematic Analsis Framework used in the Literature Review.

2.3. Data Analysis

The extracted data were subjected to qualitative synthesis in three stages:

1. Information extraction: key parameters such as electrochemical performance, degradation mechanisms, and mitigation strategies were systematically compiled.
2. Thematic synthesis: the results were clustered into the four primary strategic categories, enabling a structured evaluation of how each approach addresses specific degradation pathways in NCM.
3. Critical evaluation — the strengths, limitations, and feasibility of each strategy were assessed, providing a balanced perspective on current progress and future research opportunities.

This methodological framework ensures a systematic, comprehensive, and critical integration of diverse findings, thereby offering a consolidated understanding of advanced strategies to overcome the persistent issues in NCM cathode materials, as shown in Fig. 2.

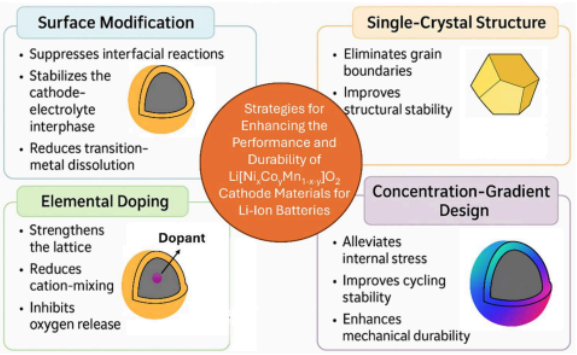

Fig. 2 Schematic Digram the strategies for Engancing Performance of NCM cathode materials.

3. Results and Discussion

A range of modification strategies has been proposed to suppress degradation phenomena in NCM cathodes (Zhang et al. 2023). Among the most widely applied are surface coatings, elemental substitution, and the development of single-crystal structures (Q. Wu et al. 2020). These methods not only extend the cycling stability of the electrodes but also provide valuable understanding of the underlying degradation pathways. For instance, lattice doping has been shown to enhance structural robustness and reduce phase transitions, while protective coatings minimize surface deterioration and suppress adverse electrolyte interactions, both of which contribute to improved overall battery stability (Li et al. 2024).

The overarching aim of these modifications is to enhance their stability during electrode production and battery cycling, enabling high-energy materials like NCM811, NCM83, and Li-rich NCM to deliver reliable cycle life and performance (Liu et al. 2021). This is particularly significant since Ni-rich NCM cathodes often suffer from limited cycle life, restricting their commercial viability. Each modification strategy targets distinct degradation routes, allowing for precise and optimized improvements in the long-term durability of the materials (Lüther et al. 2024).

3.1.Progresses in the Surface Modification of NCM

Modification of NCM cathode surfaces is commonly achieved by applying a protective layer either on individual particles or entire electrodes or by doping the surface region with foreign ions. A variety of synthesis routes have been explored, including atomic layer deposition (ALD) (Laskar et al. 2017), chemical vapor deposition (CVD) (Xu et al. 2022), magnetron sputtering (Tian et al. 2015), and wet-chemical methods (Zhu et al. 2019). Among these, ALD and CVD typically yield coatings of superior homogeneity, while wet-chemical techniques are favored for large-scale implementation due to their lower cost, despite producing comparatively less uniform layers .

The principal role of surface coatings is to form a chemically and thermodynamically stable barrier on the cathode, thereby reducing interfacial resistance and preserving structural integrity (Liu and Zou 2020). Such protective layers mitigate oxygen release, suppress transition-metal dissolution, and inhibit undesirable phase transformations, while also preventing parasitic reactions and limiting cathode–electrolyte interphase (CEI) formation. Coating materials span a wide range, including electronically conductive carbons, metal oxides (e.g., Al_2O_3, ZnO, TiO_2, MgO) (Cheng, Sivonxay, and Persson 2020; Lei et al. 2018), rare-earth oxides, lithium-containing ion conductors (e.g., Li_3PO_4, LiF, glassy phosphates) (Bandpey et al. 2023), and organic polymers (Li et al. 2023), which can be engineered in either crystalline or amorphous states depending on functional requirements.

Classification of coatings is generally based on their electrical and ionic conductivity (Tang et al. 2023). Even materials with inherently low conductivity are employed when they confer other

benefits such as high mechanical robustness, HF scavenging ability, or superior chemical stability, highlighting the multifunctional role of coatings in stabilizing Ni-rich cathodes (Q. Wu et al. 2020).

3.1.1 Electronically Conductive Coatings

Carbon-based coatings, such as amorphous carbon , graphitic carbon, and graphene oxide have long been employed to improve electronic conductivity while simultaneously protecting the surface from degradation phenomena, including HF-induced corrosion and Mn dissolution (Kucinskis, Bajars, and Kleperis 2013; Park et al. 2010). In addition, conductive polymers such as polypyrrole provide ultrathin and mechanically flexible layers that not only mitigate parasitic interfacial reactions but also accommodate the lattice strain associated with the expansion–contraction behavior of NCM cathodes during cycling (Zhang et al. 2022).

3.1.2 Ionically Conductive Coatings

Li^+ ion conductivity is essential for maintaining rate capability and capacity of NCM cathodes. The application of thin lithium-containing coatings, such as Li_3PO_4, improves the structural integrity of NCM cathodes by protecting the surface from parasitic electrolyte reactions, while simultaneously promoting Li^+ transport due to their inherent ionic conductivity (Bandpey et al. 2023; Liu et al. 2024). However, excessive coating thickness can obstruct electronic pathways and consequently reduce capacity. Thin Li-containing coatings, such as Li_3PO_4, improve stability by protecting cathode particles from electrolyte reactions and enhances Li^+ diffusion due to being good Li ion conductor. However, overly thick coatings can reduce capacity by hindering electron transport. Wet-chemical synthesis is commonly used to produce Li_3PO_4 coatings, improving both Li^+ diffusion and cycling performance (Tang et al. 2023).

3.1.3 Non-Conductive Coatings

Non-conductive coatings strengthen both the mechanical and chemical stability of cathode materials by suppressing surface degradation. Nevertheless, if the coating becomes excessively thick, it can hinder electronic and ionic transport, thereby necessitating a careful balance between protective efficiency and electrochemical performance. For example, Al_2O_3 layers have been shown to enhance cycling durability but often lead to a reduction in the initial discharge capacity (Lei et al. 2018). Analogous to Al_2O_3, TiO_2 coatings protect NCM cathodes by suppressing parasitic reactions with the electrolyte and improving long-term cycling stability. Previous studies reported that although TiO_2-coated NCM exhibited a decline in initial capacity, the overall durability during cycling was markedly improved (Fan et al. 2020).

It has been reported that, when the coating possesses higher mechanical strength than the cathode particle itself, for instance, amorphous Al_2O_3 with a hardness of up to 13 GPa (Aarik et al. 2022) compared to 11.2 GPa for NCM111 (Cheng et al. 2017), it can effectively reinforce the mechanical integrity of the electrode. Such coatings help mitigate particle volume fluctuations and delay microcrack initiation. However, to preserve electrochemical performance, the coating must allow efficient lithium-ion and electron transport to avoid impedance growth. In addition, an ideal coating material should be both cost-efficient and abundantly available. Therefore, optimizing the thickness of non-conductive coatings is essential to achieving an equilibrium between structural protection and conductive functionality.

3.1.4 Additional Effects of Coatings

In addition to their primary protective functions, surface coatings provide two less commonly highlighted benefits: broadening the electrolyte's electrochemical stability window and facilitating Li-ion transport through interfacial effects. The first mechanism involves the cathode–electrolyte interphase (CEI), which may be intentionally introduced as a coating or spontaneously generated during cycling. The CEI minimizes the mismatch in lithium chemical potential between the electrode and electrolyte at their interface, producing an additional potential drop across the thin interphase layer and thereby extending the electrolyte's electrochemical window (Zhu, He, and Mo 2015).

The second mechanism arises from the formation of a space-charge layer, which develops when two materials with different chemical potentials are brought into contact and charge neutrality cannot be maintained by electron or ion migration alone. This interfacial region exerts a strong influence on Li-ion diffusion and overall ionic conductivity. Depending on the system, the space-charge effect can enhance ion mobility in solid–solid composites or even provide extra mass storage, but it may also hinder conductivity and degrade cell performance. Although its magnitude ranges from minimal to highly significant, the space-charge layer remains a key factor in the rational design of surface coatings (Cheng et al. n.d.).

3.1.5 Coating and Doping

Chemical coating techniques typically involve a final calcination step to form the targeted protective layer on the cathode surface. For instance, investigations on ZrO_2-coated NCM have shown that annealing above 700 °C promotes Zr^{4+} diffusion into the near-surface region of the bulk, effectively resulting in partial doping of the outer lattice (Schipper et al. 2018). In contrast, Al_2O_3 coatings exhibit no detectable Al^{3+} penetration into the NCM bulk, even at annealing temperatures up to 800 °C, which is attributed to the magnetic coupling of transition metals that inhibits aluminium migration (Han et al. 2017).

A thorough analysis of the distribution of coating elements within cathode particles is crucial to distinguish between the effects of coating alone and those resulting from coating combined with doping. Coating is widely recognized for providing surface protection, whereas doping contributes to lattice stabilization within the bulk (Zhang et al. 2023). Prior studies confirm that employing both strategies can deliver complementary improvements, offering simultaneous protection at the surface and stabilization of the bulk structure. However, these modifications are often implemented through separate processes or require distinct precursors for coating and doping. Developing a single modification pathway capable of integrating both effects using the same precursor would enhance overall efficiency by reducing time and cost while achieving the desired improvements in structural integrity and surface stability.

3.2.Progresses Doping Modification of NCM

Ionic doping has been extensively employed to enhance the structural robustness of cathode materials while simultaneously improving their safety performances (Meng, Qu, and Huang 2023; Xu et al. 2024). The primary mechanisms underlying this strategy include (Mohamed and Allam 2020; Tao et al. 2021): (1) promoting Li^+ diffusion by enlarging the interlayer spacing through dopant incorporation, (2) suppressing Ni^{2+} migration into the lithium layers, thereby reducing cation mixing and maintaining structural order, and (3) mitigating oxygen release by strengthening TM–O bonding and inhibiting unfavorable phase transitions during cycling. Because the performance of ionic doping is strongly influenced by the selection of dopant species, doping techniques, and concentration levels, these parameters must be optimized to maximize the efficiency of high-Ni NCM cathodes. In practice, cation doping typically substitutes either transition-metal or lithium sites, whereas anion doping replaces oxygen atoms, both approaches aiming to stabilize the lattice and limit cation disorder. Various Dopants for $Li[Ni_xCo_yMn_{1-x-y}]O_2$ layered oxides Cathode Materials are presented in Table 1.

Table 1. Various Dopants for $Li[Ni_xCo_yMn_{1-x-y}]O_2$ layered oxides Cathode Materials

Dopant	Group	Effects on NCM Performance	Mechanism	Ref.
Y (Yttrium)	Transition Metal, Group 3	↑ Cyclability, ↑ Rate capability, ↑ Capacity retention	Larger ionic radius (Y^{3+}), reduces cation mixing.	(Kalaiselvi and Kalaignan 2019; Kang et al. 2014; Wang et al. 2010)
Ti (Titanium)	Transition Metal, Group 4	↑ Cycle life, ↑ Thermal stability, ↑ Capacity retention	Expands Li^+ transport channels, suppresses microcracks, stabilizes Ni^{2+} migration.	(Du et al. 2015; Song et al. 2018; Sun et al. 2019)
Zr (Zirconium)	Transition Metal, Group 4	↑ Cyclability, ↑ Rate capability, ↑ Thermal stability	Enlarges lattice spacing, reduces TM migration, minimizes oxygen release.	(Ding et al. 2011; Du et al. 2015; Lin et al. 2007)
V (Vanadium)	Transition Metal, Group 5	↑ Structural stability, ↑ Li-ion diffusion, ↑ Rate capability	Multi-valence states stabilize *c*-axis lattice.	(C. Lu et al. 2016; Sim et al. 2019)
Nb (Niobium)	Transition Metal, Group 5	↑ Rate capability, ↑ Cycle stability	Strong Nb–O bonds, reduces cation mixing, increases inter-slab spacing.	(Li et al. 2020; Yang et al. 2017)

Ta (Tantalum)	Transition Metal, Group 5	↑ Cyclability, ↑ Li-ion diffusion	High valence & strong Ta–O bonds	(Hendri et al. 2024; Yang et al. 2022)
Cr (Chromium)	Transition Metal, Group 6	↑ Cycling stability, ↑ Rate performance	Reduces cation mixing, stabilizes structure.	(Laine et al. 2024; Li et al. 2012)
Mo (Molybdenum)	Transition Metal, Group 6	↑ Capacity, ↑ Thermal & cycle stability	strong Mo–O bonds; stabilizes phase, segregates at grain boundaries; suppresses O release.	(Breuer et al. 2018; Sattar et al. 2020)
W (Tungsten)	Transition Metal, Group 6	↑ Cycling stability, ↑ Thermal stability	Strong W–O bonds, stabilizes layered structure, good at high cutoff voltages (4.5 V).	(Geng et al. 2022; Kim et al. 2018)
Fe (Iron)	Transition Metal, Group 8	↑ Capacity, ↑ cycle stability	Superior structural integrity. Co-doping (Fe/Al, Ti/Fe) improves rate & structure.	(M. Lu et al. 2016; Park et al. 2019)
Cu (Copper)	Transition Metal, Group 11	↑ Li-ion diffusion, ↑ Conductivity, ↑ Capacity retention	Optimal 2 mol% improves cycling stability; reduces cation mixing in Li-rich NCM.	(Milewska and Molenda 2012)
Au (Gold)	Transition Metal, Group 11	↑ Cyclability, ↑ Rate capability	Au nanofibers (2 mol%) improve Li-ion pathways & reduce resistance.	(Yue et al. 2018)
Zn (Zinc)	Transition Metal, Group 12	↑ Cyclability, ↑ Rate capability (but ↓ initial capacity at high content)	Expands lattice spacing; excess Zn → capacity loss. Optimal ~2 mol%.	(Yue et al. 2018)
Cd (Cadmium)	Transition Metal, Group 12	↑ Crystallinity, ↑ Cycling stability	Strong Cd–O bond, increases lattice parameters	(Dong et al. 2019)
Na (Sodium)	Alkali Metal	↑ Lattice expansion, ↑ Rate capability	<3 mol% improves cyclability; excess → void formation.	(Yue et al. 2021)
K (Potassium)	Alkali Metal	↑ Structural stability, ↑ Layer spacing	Acts as "pillar" in Li-layer, reduces cation mixing; excess → lattice distortion.	(Huang et al. 2023; Mo et al. 2020)
Rb (Rubidium)	Alkali Metal	↑ Capacity retention, ↑ Cycling stability	Suppresses O_2 loss, but excessive Rb → incomplete substitution.	(He et al. 2019; Li et al. 2017)
Mg (Magnesium)	Alkaline Earth Metal	↑ Structural stability, ↑ Capacity retention	Strong Mg–O bonds, reduces resistance, mitigates collapse in NCM811.	(Cho et al. 2017; Ma et al. 2025)
Ca (Calcium)	Alkaline Earth Metal	↑ Stability, ↑ Rate performance	Pillar effect; large ionic radius may cause strain. Optimal ~4.8 mol%.	(Wang et al. 2021)
Ba (Barium)	Alkaline Earth Metal	↑ Conductivity, ↑ Cycling performance	Non-homogeneous distribution; optimal ~8 mol%.	(Wang et al. 2021)

Al (Aluminum)	Post-transition, Group 13	↑ Structural stability, ↑ Cycle life	Strong Al–O bonds; suppresses lattice contraction & microcracks.	(Dang et al. 2022; Jeon et al. 2022)
Ga (Gallium)	Post-transition, Group 13	↑ Cyclability, ↑ Li-ion transport	Expands lattice; Ga/Zr co-doping further boosts kinetics.	(L. Wu et al. 2020)
Sn (Tin)	Post-transition, Group 14	↑ Stability, ↑ Rate performance	Strong Sn–O bonds; excessive Sn → secondary phase & disorder.	(Nguyen et al. 2021)
Pb (Lead)	Post-transition, Group 14	↑ Stability, ↑ Cycle life	Prevents oxygen release in Li-rich NCM; excess → impurities. Optimal ~2 mol%.	(Zhang et al. 2019)
B (Boron)	Metalloid	↑ Stability, ↑ Li-ion migration, ↓ Cracks	Expands cell volume, suppresses rock-salt phase.	(Roitzheim et al. 2022)
Si (Silicon)	Metalloid	↑ Reversible capacity, ↑ Stability	Expands lattice, suppresses phase transitions; effective in Li-rich NCM.	(Weigel et al. 2019)
Ge (Germanium)	Metalloid	↑ Structural stability	Mostly theoretical studies; suppresses O_2 release, lattice distortion.	(Chen et al. 2021)
Sb (Antimony)	Metalloid	↑ Surface stability, ↑ Cycling	0.1–0.3 mol% beneficial; >0.5 mol% → Sb_2O_3 secondary phase forms coating.	(Li et al. 2019)
La (Lanthanum)	Lanthanide	↑ Stability, ↑ Capacity	Prevents surface degradation in Li-rich NCM.	(Ding et al. 2007; DONG et al. 2017)
Ce (Cerium)	Lanthanide	↑ Stability, ↑ Surface protection	Strong Ce–O bonds;	(ZHONG et al. 2011)
F (Fluorine)	Nonmetal	↑ Stability, ↑ Cycle life	Forms Li–F bonds, enlarges Li-layer spacing, resists HF attack.	(Zhang, Duan, and Zhang 2021)
P (Phosphorus)	Nonmetal	↑ Structural stability	Forms strong PO_4^{3-} bonds	(Yuan et al. 2020)
S (Sulfur)	Nonmetal	↑ Capacity, ↑ Stability	Suppresses phase transitions; effective in Li-rich NCM.	(Lee et al. 2021)
Cl (Chlorine)	Nonmetal	↑ Li-ion diffusion, ↑ Stability	Enlarges Li-layer spacing; co-doping with Br/K effective.	(Hai-Lang Zhang 2013)
Br (Bromine)	Nonmetal	↑ Conductivity, ↑ Stability	Improves ionic/electronic transport; co-doping with Na/Cl effective.	(Zhu et al. 2018)

3.3.Single Crystal Structure

Single-crystal NCM (SC-NCM) cathodes have gained considerable interest in both academic and industrial fields due to their distinct advantages over polycrystalline NCM (PC-NCM) counterparts (Tao et al. 2021). Key benefits include superior structural integrity, enhanced cycling stability, higher thermal tolerance, and suitability for operation at elevated voltages. Unlike

polycrystalline materials, SC-NCM lacks grain boundaries, thereby reducing crack formation. Furthermore, its relatively lower surface area minimizes undesirable interfacial side reactions, phase transformations, and gas evolution, contributing to improved safety and reliability under high-temperature and high-voltage conditions (Park et al. 2018).

Despite these advantages, SC-NCM still encounters several challenges (Li et al. 2022). A primary concern is the reduced lithium-ion diffusion rate within the single-crystal lattice, which can lead to inhomogeneous Li distribution, internal stress buildup, and eventual crack initiation. Fabrication of SC-NCM is also complex and costly, with current high-temperature synthesis processes prone to lithium–nickel cation intermixing. In addition, SC-NCM, similar to other NCM systems, remains vulnerable to issues such as transition-metal dissolution and surface reconstruction.

To address these limitations, various approaches have been proposed, including crystal facet engineering, grain size optimization, elemental doping, and surface/interface modification. Studies indicate that the redox activity of SC-NCM is highly dependent on the state of charge (SOC), with performance markedly enhanced at high SOC when coupled with surface or subsurface modifications. Innovative architectures, such as radially aligned micro-sized particles exposing active crystal planes have demonstrated improved Li-ion transport kinetics, reduced mechanical stress, and superior electrochemical characteristics, including higher capacity and better rate capability. Moreover, the single-crystal morphology enhances thermal stability by mitigating parasitic reactions, thereby ensuring reliable performance under demanding operating conditions (Li et al. 2024).

Comparative investigations further confirm that SC-NCM exhibits stronger resistance to oxygen release, superior cycling durability, and higher energy density compared to polycrystalline analogues. Its long-term stability is particularly notable, with only minor performance degradation over extended cycling. Current research continues to focus on strategies such as heteroatom doping, protective surface coatings, and tailored electrolyte additives to further optimize SC-NCM for high-voltage and high-temperature applications. Nonetheless, substantial progress in processing techniques and cost reduction remains essential before SC-NCM can achieve widespread commercial adoption (Chen et al. 2024).

3.4.Concentration Gradient Structure

When the nickel content exceeds 0.8, the cathode–electrolyte interfacial reactions become unstable, accompanied by pronounced structural contraction during cycling. To mitigate these issues, strategies have been proposed to lower the surface Ni concentration in cathode particles, thereby enhancing their cycling stability (Tao et al. 2021). Three representative structural designs have emerged from this approach: core–shell (CS), core–shell gradient (CSG), and full-concentration gradient (FCG) architectures. These configurations are intended to reinforce structural stability, suppress parasitic interfacial reactions, and alleviate detrimental volume changes.

3.4.1Core-shell Structure (CS) Cathode: Reduce the Internal Stress Concentration

Sun et al. (Sun et al. 2005) introduced a core–shell cathode architecture consisting of a high-capacity Ni-rich core encased within a structurally stable shell, as exemplified by $Li[(Ni_{0.8}Co_{0.1}Mn_{0.1})_{0.8}(Ni_{0.5}Mn_{0.5})_{0.2}]O_2$. This configuration delivered a discharge capacity of 188 mAh g^{-1} within the potential range of 3.0–4.3 V. The shell exerts compressive stress on the Ni-rich core, effectively counterbalancing the tensile stress generated during phase transitions, thereby reducing localized stress accumulation and slowing structural deterioration. Nonetheless, the core experiences more pronounced phase transitions than the shell, creating substantial radial stress at the core–shell interface, which may lead to interfacial debonding. Wu et al. (Wu and Lu 2017) further reported that optimal structural compatibility requires maintaining shell thickness below 0.7 μm, and when the shell is thinner than 0.18 μm, the core radius should not exceed 5 μm to preserve stability and mechanical integrity.

3.4.2Core-shell Gradient (CSG): Reduce Stress Concentration in the Shell

The introduction of concentration gradients in cathode materials generates a distinctive radial crystallographic texture that overcomes the limitations of conventional designs. Sun et al. (Sun et al. 2005, 2009) proposed Core-shell gradient (CSG) materials with a gradient in the shell, which smooths stress transitions, prevents core-shell debonding, and enhances cyclic stability. Kim et al. (Kim et al. 2019) investigated a $Li[Ni_{0.9}Co_{0.05}Mn_{0.05}]O_2$ cathode composed of a $Li[Ni_{0.94}Co_{0.038}Mn_{0.022}]O_2$ core surrounded by a 1.5 μm CG shell with a surface composition of $Li[Ni_{0.841}Co_{0.077}Mn_{0.082}]O_2$. Their findings demonstrated a gradual phase transition that extended throughout the charging process, in contrast to the abrupt H2→H3 transition characteristic of

$Li[Ni_{0.9}Co_{0.05}Mn_{0.05}]O_2$ (CS90), thereby stabilizing the *c*-axis lattice parameter. Furthermore, Kim et al. (Kim et al. 2019) quantified the local compressive stress field arising from anisotropic volume variations. Simulation results indicated that the confining shell enhanced compressive stress in CSG90 particles and promoted a more uniform stress distribution, effectively inhibiting crack propagation. This uniformity is attributed to the elongated, rod-like grains and radially aligned primary particles inherent to CG structures, which help distribute internal forces and block electrolyte infiltration, thereby enhancing mechanical durability.

3.4.3Full-concentration Gradient (FCG) Cathode: Eliminate Stress Concentration at the Interface of the Core and Shell

Although core–shell gradient (CSG) cathodes exhibit better mechanical robustness than conventional core–shell (CS) structures, the shell thickness is sometimes inadequate to fully preserve structural stability. To overcome this drawback, the full concentration gradient (FCG) design was developed. Sun et al. (Sun et al. 2012) proposed an FCG architecture in which the nickel concentration gradually decreases while the manganese concentration increases from the core toward the particle surface. The FCG-NCM811 cathode showed remarkable cycling performance, maintaining nearly 85% of its capacity after 100 cycles and delivering an initial discharge capacity of 203 mAh g^{-1} (Sun et al. 2012). These findings highlight that the CSG approach significantly improves both the electrochemical capacity and long-term stability of cathode materials.

4. Conclusion

Ni-rich $Li[Ni_xCo_yMn_{1-x-y}]O_2$ cathodes offer exceptional energy density, Positioning them as strong contenders for next-generation lithium-ion batteries. However, their commercialization remains limited by degradation mechanisms such as cation mixing, oxygen release, surface reconstruction, and particle cracking, which impair stability and cycle life. This review highlighted four advanced strategies to address these challenges: surface modification, elemental doping, single-crystal engineering, and concentration-gradient architectures. Collectively, these approaches enhance structural integrity, suppress interfacial degradation, and extend electrochemical durability. Despite notable progress, critical issues remain, including capacity–stability trade-offs, interfacial instability at elevated voltages, and the absence of scalable, cost-effective synthesis methods. Future research must therefore integrate multiple strategies within unified design frameworks while emphasizing industrial scalability and environmental sustainability. In conclusion, holistic material engineering that simultaneously strengthens structural, chemical, and interfacial stability is essential to unlock the full potential of Ni-rich NCM cathodes for safe, high-performance, and commercially viable lithium-ion batteries.

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