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# A Mini Review on Ni-rich Layered Oxide Cathode Materials

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*The global energy demand and increasing global warming are threats to the planet; therefore, replacing internal combustion engine vehicles to electric vehicles is prerequisite. Rechargeable batteries are key component to operate electric vehicles, among which lithium-ion batteries are governing the industry. After the successful discovery of Goodenough's* $LiCoO_2$*, layered oxides are emerging and are considered to be the next-generation cathodes. Ni-rich layered oxides are the new-generation cathode material owing to high specific capacity, high energy density, high operating voltage and low cost. Substitution of Co, Mn and Al is advantageous to compensate for the loss of the storage capacity and structural degradation. This review aims to discuss the characteristics, structure and preparation methods for nickel-rich layered oxide cathode material.*

**Keywords:** Layered oxide, Cathode materials, Nickel rich, Lithium ion batteries

## INTRODUCTION

The worldwide concerns related to global warming and excessive greenhouse gas emissions from burning fossil fuels in internal combustion engine (ICE) vehicles have taken researchers' interest. Meanwhile, the energy demand is also increasing with the rapid growth of the population and the world economy. Therefore, it now becomes a challenge for researchers to improve existing technologies, explore new energy resources, and develop a clean, green, and sustainable environment to fulfill the world's energy needs[1–4]. Renewable energy sources such as solar energy and wind energy are the focus of energy development for a clean and green environment owing to their advantages over traditional fossil fuels. However, these energy sources are not continuous and mainly dependent on government subsidies. The purchasing cost of a solar PV module has increased by 50 times since 1980 and ten times more in 2005. Similarly, wind technology also requires considerable support and grants from the government, challenging to maintain in a long-term, economically competitive environment[5]. For a clean, green and sustainable environment, the ICE vehicles should be replaced with electric vehicles (EVs) for which rechargeable batteries are the most indispensable part for the operation of electric vehicles and other energy storage systems[6–8]. The cost of an EV battery pack is projected to reach 125 US$ kW $h^{-1}$in 2022, which is beneficial for the development of electromobility[9].

In 1991, lithium-ion batteries (LIBs) with enhanced gravimetric and volumetric energy density was commercialized by Sony for its Walkman[10,11]. Subsequently, electric vehicles were fully equipped with LIBs in 2000[12]. Further, in 2015, a new series of electric car models (Tesla Model S and X, Nissan LEAF, Chevy Volt, Ford C-Max Energi, Toyota Prius Prime, Volkswagen e-Golf, BMW i3, etc.) were introduced, which gained worldwide popularity[13]. In 2017, the estimated number of EVs reached 695,000 worldwide, with a remarkably increased sale of 50% from past years. China maintains the world's largest automobile industry, with about 50% of EVs sales, which is expected to grow by 2022, as illustrated in Figure 1[14].

# OVERVIEW OF CATHODE MATERIALS FOR LITHIUM-ION BATTERIES

## Development and classification of cathode materials for lithium-ion batteries

The journey begins with developing a $TiS_2$ cathode with lithium anode by Whittingham in 1970[15]. $LiCoO_2$ (LCO), discovered in the 1980s by Goodenough, was the first commercialized layered oxide cathode material possessing a high theoretical and volumetric capacity[16,17]. Besides, Spinel $LiMn_2O_4$ (LMO) becomes another commercialized cathode due to the inexpensive manganese; though, it suffers from severe challenges[18]. $LiFePO_4$ (LFP) with olivine structure was introduced in 1996 by the University of Texas, owning good chemical and thermal stability[19,20]. Lithium titanate also gained attention in electric power trains and smart grids[21]. For commercialization, the cathode chemistries should be integrated with transition metals (Ni, Co, Mn, or Al) to prepare $LiNiCoMnO_2$ or $LiNiCoAlO_2$ due to cost-effectiveness. Cathode chemistry primarily determine the capacity of LIBs; therefore, enhancing its energy density, cycling stability, thermal stability, and cost-effectiveness is a challenge for commercialization[22]. According to their chemistries and structures, cathode materials are classified into the following categories[23]:

1) Layered transition metal oxide ($LiMO_2$) (M=transition metal) cathode material including $LiCoO_2$, $LiNiO_2$, $LiMnO_2$, and binary or ternary material $LiNi_xCo_yM_zO_2$ (1-x-y-z), (where M is Mn, Al, Mg, and other metals).

2) Spinel transition metal oxides such as lithium manganese-based material $LiMn_2O_4$ and $LiNi_{0.5}Mn_{1.5}O_4$.

3) Olivine polyanionic compounds, including transition metal phosphate ($LiFePO_4$) silicate, borate, and pyrophosphate.

4) Other types of cathode material, such as vanadium-based materials and transition metal fluorides.

### Characteristics of cathode materials for lithium-ion batteries

Recently, the researchers achieved 200-250 Wh kg$^{-1}$ of specific energy in EV batteries; however, 500 Wh g$^{-1}$ with a total mileage of 150,000 miles and a driving range of 300 miles per single charge is required to achieve within next ten years[8,12,24]. In a characteristic battery pack, 40% of the performance is dedicated to the cathode material; therefore, the active material's chemistry, structure, and electrochemical performance should be keenly optimized. The charge/ discharge voltage for a commercial lithium battery is typically in the range of 3.4-4.1 V (vs $Li^+$), and the specific discharge capacity is around 200 mAh g$^{-1}$. To improve the performance of LIBs, the positive active material should have a high specific capacity, high energy and volumetric density, high operating voltage, superior thermal stability, high tap density, low cost, and eco-friendliness[25,26]. To achieve high performance from the cathode chemistries along with low cost, the cathode material should have the following characteristics[27]:

1) The structural stability is high. During the charge/discharge process, the lattice expansion and shrinkage are less convenient to insert/extract more $Li^+$ions.

2) The material is an excellent ionic and electronic conductor with less impedance at the cathode-electrolyte interface.

3) Redox potential is high and stable during the charge/discharge process.

4) Superior thermal stability.

5) They were synthesized using abundant raw materials with low cost and environmental friendliness.

6) The synthesis process should be facile and have a good yield for large scale production.

# Ni-RICH LAYERED OXIDE CATHODE MATERIALS

Over the last decade, Nickel-based layered oxide materials, such as $LiNi_xCo_yMn_zO_2$ and $LiNi_xCo_yAl_zO_2$ (x + y + z = 1), were considered the most suitable cathodes for LIBs due to their high specific capacity, high energy density, high operating voltage range, and low cost[28,29]. The milestone of 300 Wh g$^{\&\#8209;1}$was accomplished by Contemporary Amperex Technology (CATL) (NCM-811, pouch cell), and Panasonic used NCA, 21700 cylindrical cells[30]. In 2017, China consumed about 72% of EV industry requirements from NCM and NCA, which increased to 90% in 2018[31].

## Structural characteristics of Ni-rich cathode material

$LiNiO_2$ (LNO) was first discovered in 1954 by Dyer's group but after $LiCoO_2$ was promoted for LIBs, LNO developed as an emergent cathode due to the low cost of Ni compared to Co[32]. $LiNiO_2$ acquire $\alpha$-$NaFeO_2$ (space

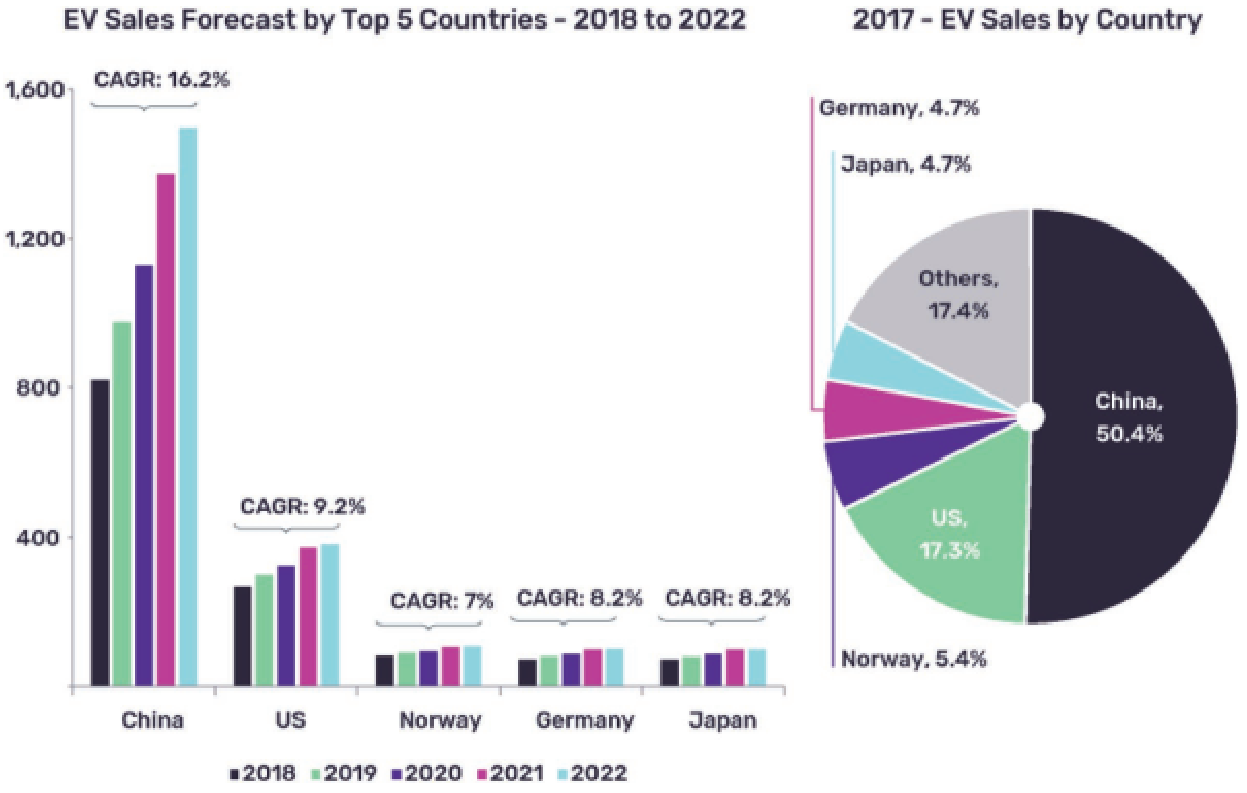

*Figure 1. EVs sales for the top five countries in the world*[14]

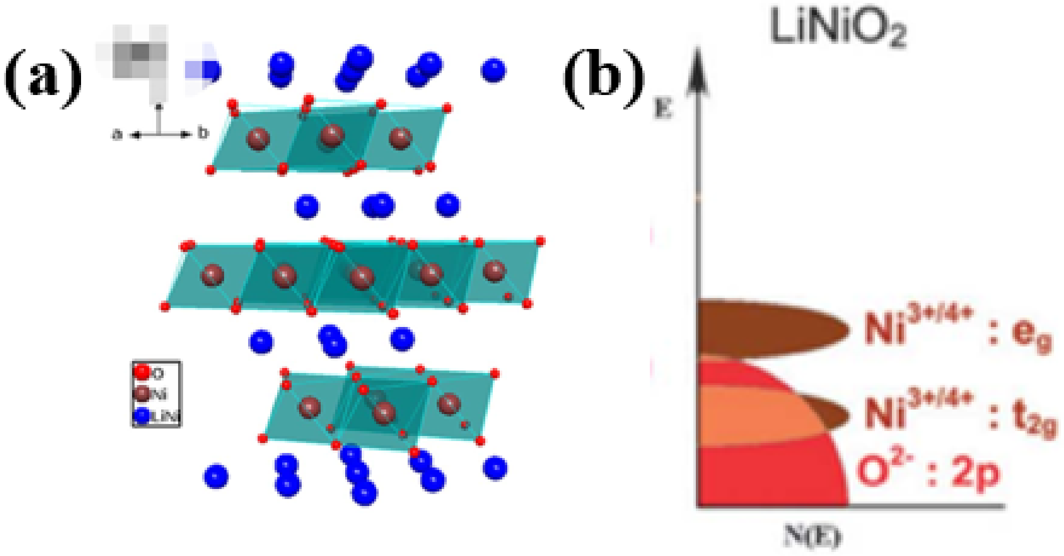

*Figure 2. (a) Crystal structure and (b) Energy diagram of* $LiNiO_2$*. Produced with permission* [33]*. Copyright 2017, Elsevier.*

group $R\bar{3}m$) structure forming a closed cubic arrangement with oxide ions and $Li^+$ and $Ni^{3+}$ employing the octahedral sites with O3 type layered structure.

The strong Ni-O-Ni covalency in low spin $Ni^{3+}$:$t2g^6$ $eg^1$ electronic configuration leads to high conductivity with the rhombohedral crystal structure by sharing their edges to form $NiO_6$ octahedron of trigonal symmetry. Nevertheless, the two-dimensional pathway for lithium diffusion is suitable for lithium conductivity (Figure **2**(a)). Moreover, the eg band of $Ni^{3+/4+}$ and $O^{2-}$:2p band are non-overlapping, enabling the extraction of one $Li^+$ per Ni ion without oxygen loss from the lattice structure as shown in Figure **2**(b)[33,34]. The cell parameters of $LiNiO_2$ (a=2.9 Å, c=14.2 Å with c/a=4.9) are equivalent to the cubic lattice, making the displacement of Li and Ni ions easier compared to $LiCoO_2$ [29].

During high-temperature calcination (about 700 °C), due to the volatilization of lithium, the structure becomes lithium deficient ($Li_{1-x}Ni_{1+x}O_2$) with Ni occupying some of the sites of lithium and the difficulty to stabilize all the Ni ions in $Ni^{3+}$ valence state due to the partial reduction of $Ni^{3+}$ to $Ni^{2+}$ will eventually result in low rate capability[35,36]. Ohzuku et al. [37] reported that the peak intensity ratio of I (003)/I (104) could be a prime factor to demonstrate the degree of cation mixing and can ultimately predict the electrochemical performance of $LiNiO_2$. However, during the charge/discharge process, $LiNiO_2$ also endures the phase transition, which leads to capacity decay. During the 1*st* charge cycle, the $Ni^{2+}$ present in the TM layer gets oxidized; however, with further lithium extraction, $Ni^{2+}$ in the lithium layer also gets oxidized, resulting in the inter-slab layer contraction[38]. Dahn et al.[39] reported that the electrochemical insertion of additional $Li^+$ into $LiNiO_2$ forms a new $Li_2NiO_2$ structure. Thus, during the removal of additional $Li^+$, the redox potential becomes comparatively low resulting in poor structural stability. To resolve the problems of non-stoichiometric $Li_{1-x}Ni_{1+x}O_2$, substitution with Co, Mn, and Al are beneficial, which is discussed in the following section.

$LiNiO_2$ is isostructural to $LiCoO_2$, owing to the similar ionic radius of $Ni^{3+}$(0.56 Å) with $Co^{3+}$ (0.545 Å) and the similar electronegativity of Ni (1.91) and Co (1.88) as well as both cathodes require oxygen flow during the synthesis process. Hence, it is beneficial to substitute Ni with Co to minimize cation mixing and suppress the phase transition. The amount of $Ni^{2+}$ reduces when the substitution of cobalt increases in $LiNi_{1-x}Co_xO_2$, and finally, a stoichiometric $LiNi_{0.7}Co_{0.3}O_2$ is synthesized without oxygen flow as reported by Rougier's group[40].

## Methods for the preparation of Ni-rich layered oxide cathode material

In general, the preparation method of Ni-rich cathodes including solvothermal method [46], co-precipitation method[47], sol-gel method[48], high-temperature solid-state route[49], sublimation-induced gas-reacting (SIGR) process[50], molten salt method[51], microwave-assisted co-precipitation method[52], spray drying process[53], etc. Among all the synthesis methods, the co-precipitation method can precisely control the morphology at a microscopic level and maintain the elemental distribution. It is also beneficial for large-scale production for industrialization. The preparation steps include mixing the solutions of transition metals in a continuous stirring tank reactor (CSTR) with the precipitator (NaOH is generally preferred). The nucleation process is very high, which results in the formation of microscopic-sized irregular grains. The composition of transition metal elements is designed according to the requirement. In order to obtain the higher density and particle size being uniformly distributed, a chelating agent is required, such as ammonia solution while carefully adjusting the concentration of the chelating agent, pH value (10.5-11.5), reaction temperature (40-60 ºC), stirring rate, feeding rate, and other parameters is crucial. After continuously stirring in a tank reactor for hours, the particles steadily grow and agglomerates to give the desired morphology and elemental composition. The product obtained is a precursor that is further washed, filtered, and dried to obtained Ni-based cathode material. The precursor is further mixed with a lithium source (specifically LiOH and $Li_2CO_3$) followed by high-temperature calcination. Liu et al.[52] successfully prepared $LiNi_{0.85}Co_{0.05}Mn_{0.1}O_2$ via microwave-assisted co-precipitation method. The advanced synthetic process provides the controlled morphology, well-ordered phase structure, high tap density, and lower cation mixing.

However, for high-Ni cathodes (NCM811), the preparation steps become more complex than the preparation process of NCM523, and the equipment should meet the general standards. They also require LiOH as a lithium source instead of $Li_2CO_3$, flowing environment should be $O_2$, long-duration for calcination, post calcination treatment, etc. The sintering environment should be strictly controlled by moisture and erosion reaction. The synthesis parameters such as pH, heating temperature, and ammonia solution should also be adjusted carefully according to the Ni content. The usage of more LiOH, calcination duration and $O_2$ flow costs more in high-Ni oxides; however, it balances the cost of cobalt in NCM523. Hence, the price of NCM811 is comparable with NCM523 however is much lower than NCM111[8,54].

## CONCLUSIONS & OUTLOOK

Rechargeable batteries are crucial for electric vehicle and other energy storage applications. Lithium-ion batter-

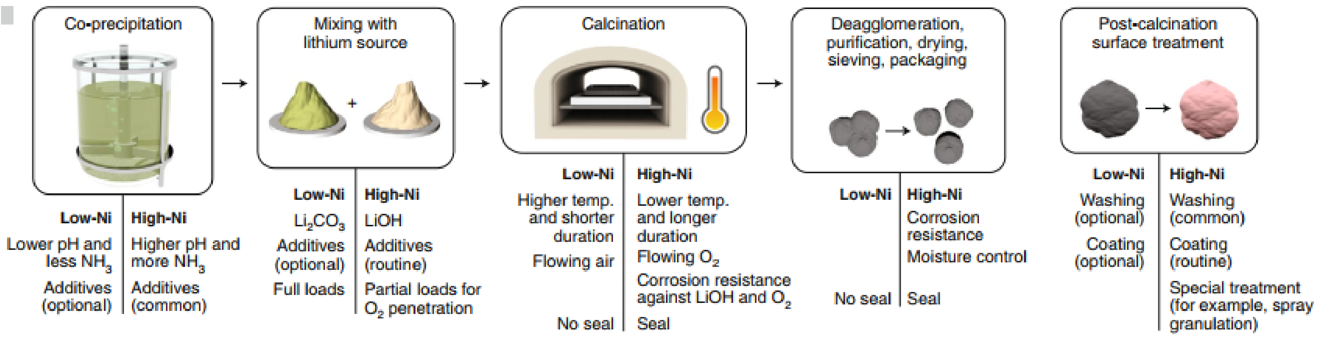

*Figure 3. Schematic illustration of manufacturing synthesis of nickel-based layered oxides. Produced with permission*[8]*. Copyright 2020, Springer Nature.*

ies are dominant and their demand is increasing with increasing energy demands. Layered oxides as cathode material are beneficial and their cathode chemistries are suitable to faciliatet high-energy-density for commercialization. Ni-rich layered oxide positive electrode material is considered as next-generation cathode material due to high discharge capacity, high working voltage, and low cost for their application in high-energy lithium-ion batteries (LIBs). Efficient and one-step synthesis strategies are the demand for industrialization of Ni-rich cathodes. One-step synthesis strategies are preferred over multiple-step strategies due to the longer synthesis duration and calcination environment, which requires $O_2$ gas. Substitution of cobalt, manganese and aluminum gives rise to $LiNi_xCo_yMn_zO_2$ (NCM) and $LiNi_xCo_yAl_zO_2$ (NCA) (x+y+z=1) that can effectively compensate for the performance loss.

Recently, researchers are focusing on reducing and eliminating cobalt from Ni-based cathodes as Co is scarce, expensive, and presents ethical issues in mining. Co-free cathodes are placing a next step towards the commercialization of Ni-rich cathodes. However, the absence of Co presents severe challenges in terms of poor cycling stability and electrochemical properties of Ni-rich layered cathodes. High-Ni (Ni> 0.9) cathodes are also a demand to fulfill the driving range of 300 miles per charge and cost-effectiveness due to the low cost of Ni. Nevertheless, Ni content plays a vital role in the structural and electrochemical stability of Ni-rich cathodes. Hence, by overcoming the material defects, the high specific capacity and high energy density of Ni-rich cathode materials can be achieved for its application in next-generation LIBs.

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