


## Review Article

# Progress in Preparation and Modification of $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ Cathode Material for High Energy Density Li-Ion Batteries

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Due to the advantages of high specific capacity, various temperatures, and low cost, layered  $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$  has become one of the potential cathode materials for lithium-ion battery. However, its application was limited by the high cation mixing degree and poor electric conductivity. In this paper, the influences of synthesis methods and modification such surface coating and doping materials on the electrochemical properties such as capacity, cycle stability, rate capability, and impedance of  $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$  cathode materials are reviewed and discussed. The confronting issues of  $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$  cathode materials have been pointed out, and the future development of its application is also prospected.

## 1. Introduction

To meet the continuously increasing demand of clean energy globally, the rechargeable Li-ion batteries have been used in various areas like electric vehicles, communications, military, energy, and other fields [1]. Cathode material has a significant impact on the electrochemical properties and safety of lithium battery. So the cathode material has a crucial role in accelerating popularization and adaptation of the Li-ion secondary battery. It is well known that  $\text{LiFePO}_4$  as the traditional lithium-ion battery cathode material has a low energy density [2];  $\text{LiCoO}_2$  has excellent electrochemical performance, but cobalt is scarce and toxic [3]; the  $\text{LiNiO}_2$  has serious cation mixing of  $\text{Ni}^{2+}$  and  $\text{Li}^+$  and high irreversible capacity [4]. Layered  $\text{LiMnO}_2$  has crystallographic transformation to spinel structure [5] and spinel  $\text{LiMn}_2\text{O}_4$  has the Jahn-Teller distortion during charging and discharging [6]. The above-mentioned cathode materials are inherently limited by their own limitations. Therefore, to develop an optimum cathode material with high energy density, long cycle life and excellent thermal stability have become a hot topic around the world.

Due to the synergistic effect of the Ni, Co, and Mn,  $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$  (NCMxyz) as a new type of energy-storage materials with high specific capacity and high capacity retention ratio has attracted much attention [7]. Particularly, the nickel-rich NCMxyz cathode materials ( $x \geq 50\%$ ) deliver high capacity such as NCM622 [8], NCM71515 [9], and NCM811 [10]. As is known, the cation mixing of  $\text{Li}^+$  (0.76 Å) and  $\text{Ni}^{2+}$  (0.69 Å) in the NCM cathode materials results in the lattice distortion and breakdown of layered structure [11]. However, the cation mixing has been proved to cause the sharp drop of energy density and structure deterioration [12]. As shown in Figure 1, when the Ni content increased, the specific discharge capacity increased, while the capacity retention and thermal stability decreased [13]. Recently, Cui et al. [14] have measured the Li-ion diffusion coefficient of NCM materials ((111), (422), (523), (525), (622), and (71515)) from  $-25$  to  $50^\circ\text{C}$  and found that the Li-ion diffusion coefficient of NCM622 was highest with the minimum temperature effect among all the NCM materials. Obviously, the NCM622 has been one of the most promising cathode materials for Li-ion batteries with excellent electrochemical properties.

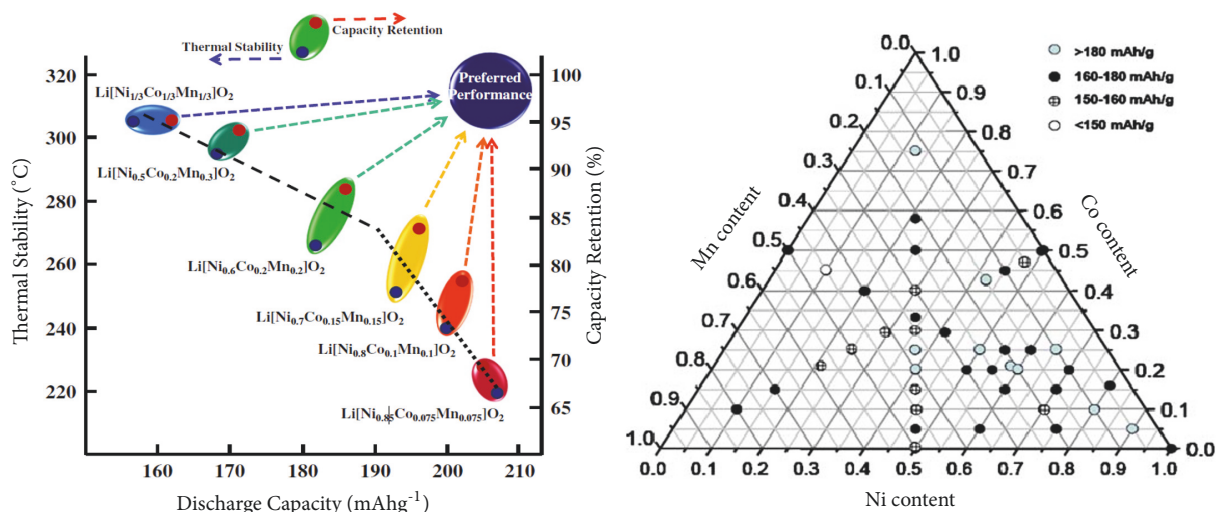


FIGURE 1: Elementary composition and electrochemical properties diagram of  $\text{Li}[\text{Ni}_x\text{Co}_y\text{Mn}_z]\text{O}_2$  [13]. Copyright: Journal of Solid State Electrochemistry, 2009, and Journal of Power Sources, 2013.

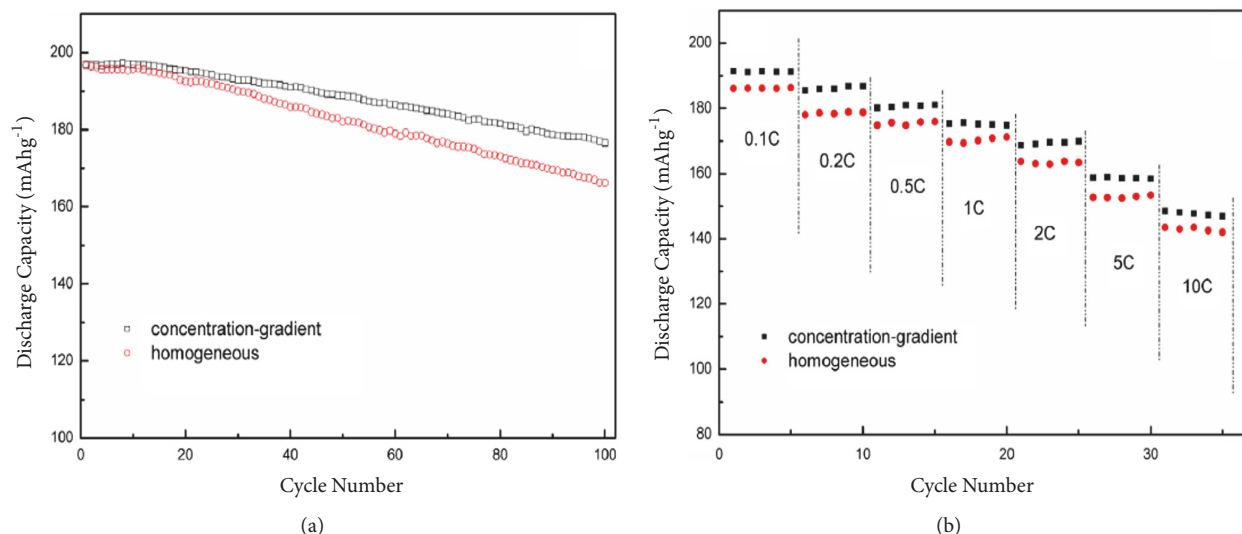


FIGURE 2: (a) Cycle performance and (b) rate capability of NCM622 with different synthetic methods [15]. Copyright: Journal of Alloys and Compounds, 2014.

## 2. Preparation of NCM622

At present, many methods have been devoted to addressing synthesis obstacles, such as the coprecipitation method, spray drying method, high temperature solid state reaction, and combustion method. Different methods have great influences on the electrochemical properties of NCM622 cathode materials.

**2.1. Coprecipitation Method.** Coprecipitation method is a useful preparation process for the industrial production of cathode materials. This method can synthesize precursor with excellent spherical morphology and element mixing at an atomic level. The precipitation conditions such as coprecipitation temperature, pH value of solution, and stirring intensity play a decisive role in the performance of precursor.

The effects of hydroxide coprecipitation conditions of  $\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}(\text{OH})_2$  were systematically studied by Liang et al. [16]. They pointed out that the particles became small with an increase in the pH value, while the particles of precursor became quasi-spherical with increasing chelating agent concentration and stirring speed. As is known, the inhomogeneous composition is a major difficulty in coprecipitation. Li et al. [17] synthesized the hydroxide precursor with different Co content using coprecipitation method. When the Co content in  $\text{Ni}_{0.6}\text{Mn}_{0.4-x}\text{Co}_x(\text{OH})_2$  increased, the tap-density and the initial discharge capacity of NCM622 increased, but their cycling stability decreased owing to the acceleration of grain growth [17]. The NCM622 cathode materials with the concentration gradient of Mn and Ni elements were synthesized using hydroxide coprecipitation method [18]. As seen in Figure 2, the electrochemical

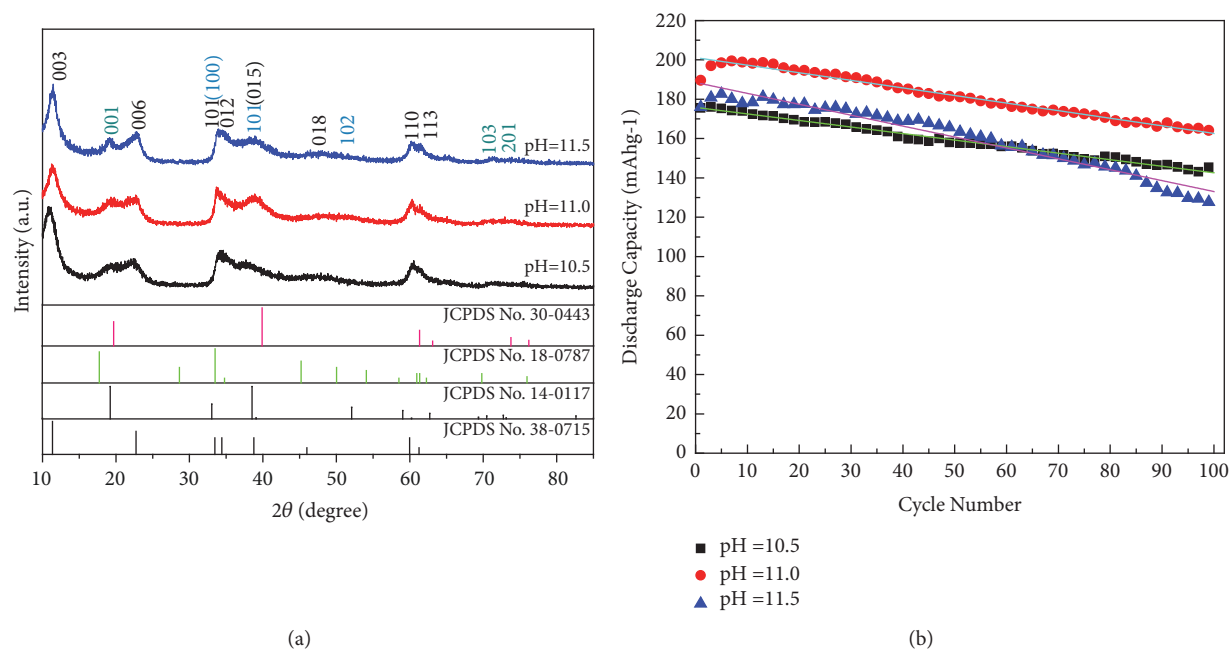


FIGURE 3: (a) XRD diffraction patterns of  $(\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2})\text{OH}_2$  and (b) cyclic performance curve of NCM622 with different pH values [19]. Copyright: Ionics, 2017.

properties of concentration-gradient cathode materials were better than those of the homogeneous cathode material. The concentration-gradient synthesis method could be effective in reducing the cation mixing in the Ni-depleted outer shell layer of cathode material [15].

The different crystals of precursor are formed under different coprecipitation conditions, such as pH values and temperatures. The  $(\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2})\text{CO}_3$  precursors were synthesized as the pH values varied from 7 to 9.5 [20]. When the pH value reached 9.5, the  $(\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2})\text{OH}_2$  phase was precipitated. But when the pH value fell to 7.0, the Mn content decreased sharply in the precipitate. As shown in Figure 3(a) [19], the  $(\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2})\text{OH}_2$  precursors with a mixed phase of  $\alpha$  and  $\beta$  were synthesized with hydroxide coprecipitation method using sodium DL-lactate as an eco-friendly chelating agent. The effects of the pH value on the precursor structure and the electrochemical properties of the cathode materials were investigated and analyzed. It was found that the NCM622 cathode material at pH=11.0 possessed the highest discharge capacity and capacity retention (Figure 3(b)).

The electrochemical properties of the cathode material are also influenced by the micromorphology of the precursor. Kim et al. [21] synthesized the NCM622 cathode materials using different inorganic salts (sulfate, nitrate, and acetate) as raw materials. The FE-SEM images showed that sulfate and acetate starting materials formed nanosized primary particles and void channels, whereas nitrate has no void channels. Sulfate with void channels and a regular layered structure displayed the best electrochemical performances. A porous microsphere NCM622 cathode material with a diameter of  $3\mu\text{m}$  was prepared using the carbonate coprecipitation method and impregnation method [22]. The rate

capability of nanocrystal-assembled porous NCM622 was improved dramatically in comparison to that of submicron-sized NCM622. This result was probably due to the porous interiors of structure. The unique structure could ensure the structural stabilization via alleviating the mechanical strain induced by volume change at a high current density.

The cathode materials are prepared by heating the precursor and lithium at high temperature in air. The cathode materials sintering at the different conditions have the different electrochemical properties. The sintering temperatures and sintering times of  $(\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2})\text{CO}_3$  precursors were studied in detail. When the optimum sintering temperature and sintering time were  $850^\circ\text{C}$  and 20 h, respectively, the corresponding cathode materials possessed the highest discharge capacity and capacity retention rate [23]. But for the  $(\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2})\text{OH}_2$  hydroxide precursor, its optimum sintering temperature was  $900^\circ\text{C}$  [24]. This indicated that the carbonate precursor has more advantages in the sintering process than hydroxide precursor.

**2.2. Spray Drying Method.** Spray drying is another useful method to synthesize the cathode materials with element mixing at the atomic level [25]. In this case, the cathode particles are tiny and quasi-spherical. Furthermore, the spray drying method can achieve continuous production of automatic control and have strong preparation ability and high production efficiency. However, the large-scale production of spray drying method is restricted owing to the complicated process and the high precision of the equipment. As shown in Figure 4, the single-phase layered NCM622 cathode materials were prepared with spray drying method using acetate as raw materials [26]. The cathode materials have very excellent crystallization performance, and the surface of

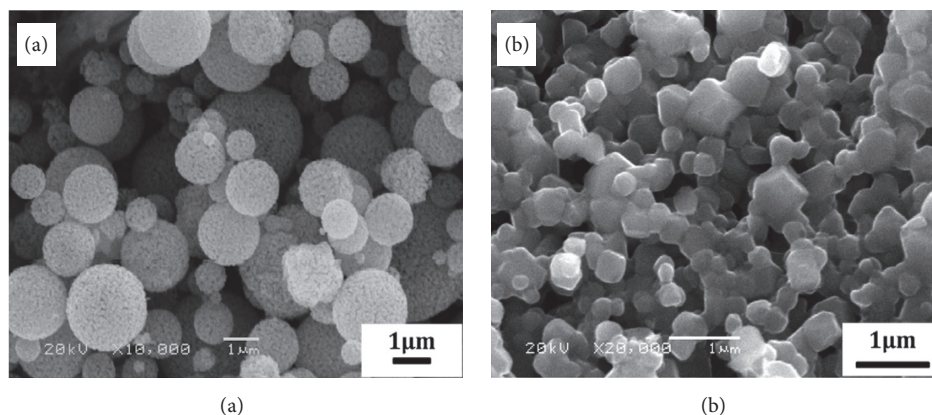


FIGURE 4: SEM of the (a) precursor powder and (b) NCM622 cathode powder prepared by ultrasonic spray drying [26]. Copyright: Materials Letters, 2015.

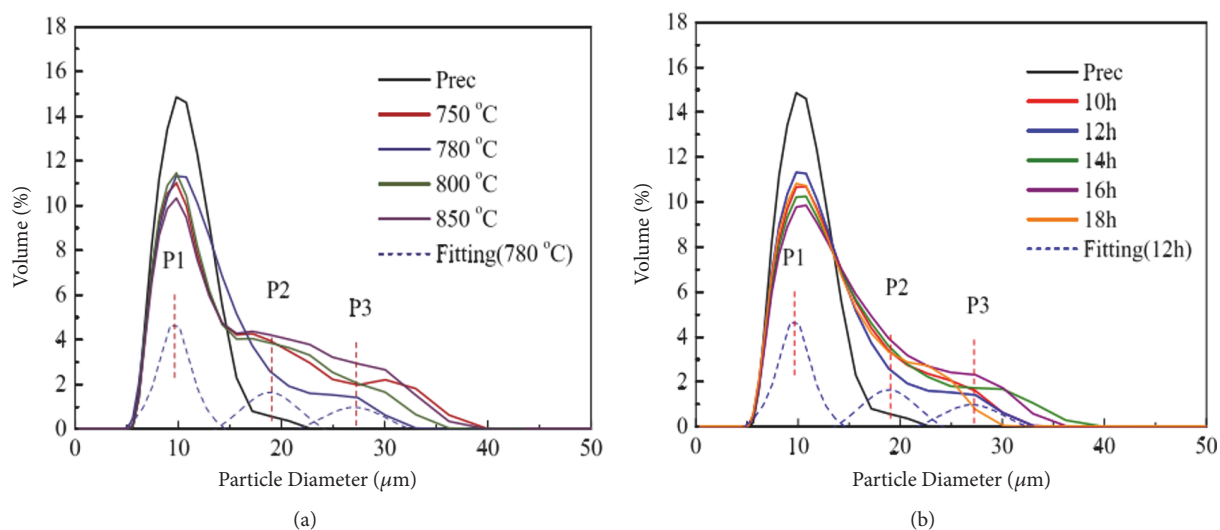


FIGURE 5: Particle size of NCM622 prepared by solid state reaction at (a) sintering temperatures and (b) sintering times [29]. Copyright: Ceramics International, 2015.

particle was smooth and quasi-spherical. Subsequently, the effects of different spray sintering temperatures (700-900 °C) on the performance of NCM622 cathode materials were systematically studied [27]. The cathode material sintered at 850 °C has excellent electrochemical performance with the initial discharge capacity of 173mAh/g and good cycling performance.

As the improvements of technology, the oxidative precursors (a hybrid of  $\text{NiO}/\text{MnCo}_2\text{O}_4/\text{Ni}_6\text{MnO}_8$ ) were prepared with ultrasonic spray thermal decomposition method [28]. As compared with the coprecipitation precursor, the oxidative precursor has little influence on the electrochemical performances of cathode material, but the preparation process of ultrasonic spray thermal decomposition method was greatly reduced.

**2.3. Solid State Method.** For solid state reaction method, the metal salt and lithium salt are measured precisely and mixed evenly, and then the cathode materials are prepared

through burning mixture directly at high temperatures. In comparison with other methods, the solid state reaction method has low requirements of equipment, and the reaction can be managed easily. However, there are technical problems such as uneven mixing of raw material and poor uniformity of particle size. In order to minimize the presence of impurity ions, the corresponding hydroxides and oxides are usually used. Due to the large segregation of compositions and the appearance of hetero phase, the cathode material prepared with solid state reaction method has poor electrochemical performance. For example, Xia et al. [29] explored the effect of the sintering conditions on electrochemical properties of NCM622 cathode materials when they used the segmented sintering solid state reaction method and indicated that a larger particle size and severer agglomeration were observed and occurred at 850 °C (Figure 5(a)). As seen in Figure 5(b), with increasing the sintering time, the primary particle size of cathode materials did not become large obviously, but their electrochemical performance reduced.

TABLE 1: Discharge capacity and fade rate of samples operated at different temperatures [30].

| Temperature | prepared method | Discharge capacity of 1C (mAh/g) | Fading rate per cycle (mAh/g) |
|-------------|-----------------|----------------------------------|-------------------------------|
| 25°C        | Spray-drying    | 143.7                            | 0.44 (0.31%)                  |
|             | Solid -state    | 138.4                            | 0.48 (0.34%)                  |
| 50°C        | Spray-drying    | 159.3                            | 0.55 (0.35%)                  |
|             | Solid -state    | 156.1                            | 0.94 (0.60%)                  |

TABLE 2: Electrochemical performance of NCM622.

| Authors      | Synthesis methods          | Testing conditions | Initial discharge capacity (mAh/g) | Capacity retention rate (%) | Ref. |
|--------------|----------------------------|--------------------|------------------------------------|-----------------------------|------|
| Liang et al. | Hydroxide co-precipitation | 1C, 2.8–4.3        | 172.1                              | 94.3 (100 cycles)           | [16] |
| Li et al.    | Hydroxide co-precipitation | 30 mA/g, 2.8–4.3   | 172.8                              | 71.8 (50 cycles)            | [17] |
| Zhang et al. | Carbonate co-precipitation | 0.2C, 2.8–4.3      | 180.0                              | 82.4(30 cycles)             | [20] |
| Xu et al.    | Hydroxide co-precipitation | 0.1C, 3.0–4.3      | 201.6                              | 90.1 (100 cycles)           | [19] |
| Zhong et al. | Carbonate co-precipitation | 0.2C, 3.0–4.3 V    | 148.0                              | 91.8 (30 cycles)            | [23] |
| Yue et al.   | Spray-drying               | 1C, 3.0–4.3 V      | 160.8                              | 93.7 (40 cycles)            | [26] |
| Yue et al.   | Spray-drying               | 80mA/g,3.0–4.3V    | 155.7                              | 89.0 (50 cycles)            | [27] |
| Li et al.    | Spray-drying               | 1C, 2.8–4.3 V      | 160.8                              | 90.8 (100 cycles)           | [28] |
| Xia et al.   | Solid state                | 1C, 2.8–4.3 V      | 156.3                              | 102.9 (100 cycles)          | [29] |
| Yue et al.   | Solid state                | 1C, 2.8–4.3 V      | 138.0                              | 82.9 (100 cycles)           | [30] |
| Ahn et al.   | Combustion                 | 0.1C, 3.0–4.3 V    | 170.0                              | 98.2 (30 cycles)            | [3]  |

The differences between solid state reaction and spray drying on the electrochemical performances of NCM622 cathode materials were studied at room and elevated temperatures [30]. As shown in Table 1, the sample with spray drying method exhibited the higher discharge capacity and better cycling performance than that with solid state method.

**2.4. Combustion Method.** In the combustion synthesis, the organic metal salts are usually first mixed with nitric acid/urea, and then the above mixture is heated directly to the ignition temperature. Finally the cathode materials are synthesized by the exothermic heat of the material in the chemical reaction. The combustion method is known for its main advantages of simple equipment, low cost, and being without external energy. But it is restricted because of the shortcomings of large particle size and poor controllability. Few researchers prepare the cathode material with this method. For example, Ahn et al. [3] prepared the precursor powders through burning the mixture of acetate and nitric acid/urea. Then, the effect of sintering temperature (700–1000°C) on the performance of the cathode material was studied. The results showed that the  $\text{Ni}^{3+}/(\text{Ni}^{2+} + \text{Ni}^{3+})$  value of 0.690 for the NCM622 sintered at 800°C was closest to the theoretical value (0.667). Meanwhile, the NCM622 prepared at 800°C showed the best layered structure with the maximum  $c/a$  value.

The discharge specific capacities and cyclic stabilities of NCM622 cathode material synthesized with different preparation methods were listed in Table 2. Among the preparation methods mentioned in Table 2, most of NCM622 cathode materials prepared with coprecipitation method displayed highest capacities. Furthermore, the coprecipitation method with the homogeneous elements mixing at the atomic level and simple experiment conditions has been regarded as the

most effective way to synthesize the precursor of NCM622 cathode materials. At present, carbonate coprecipitation process has certain advantages in terms of cost control. But the particle size of the precursor is controlled difficultly, and the tap-density of carbonate precursor is lower than that of hydroxide precursor [24]. The solubility product ( $K_{sp}$ ) of carbonate coprecipitation product is bigger than that of hydroxide. Thus, the hydroxide coprecipitation method is the most effective method to prepare the NCM622 cathode materials.

### 3. Modification of NCM622

It is imperative to further improve the electrochemical performance of cathode material for the next generation electric vehicles. The long-term cycling is directly correlated with the capacity loss. The mechanical stress heterogeneity is ultimately attributed to intergranular fracturing that degrades the connectivity of subsurface grains and causes the capacity loss [31]. As is known, when the same cathode material is modified with different modification materials, their electrochemical properties could behave with great differences [32].

**3.1. Surface Coating Method.** The capacity fading of the nickel-rich cathode material has become serious during charge-discharge. At present, the surface coating as a mainly modification method is used to improve the cycle stability and thermal stability of the cathode materials. The commonly used coating materials for cathode materials are oxides, mineral salts, and active electrode materials.

When  $\text{TiO}_2$  is used for coating, the inert  $\text{TiO}_2$  coating on the cathode surface can protect the active material from reacting with the electrolyte and inhibit the cathode materials dissolution during charge-discharge cycles. For instance,

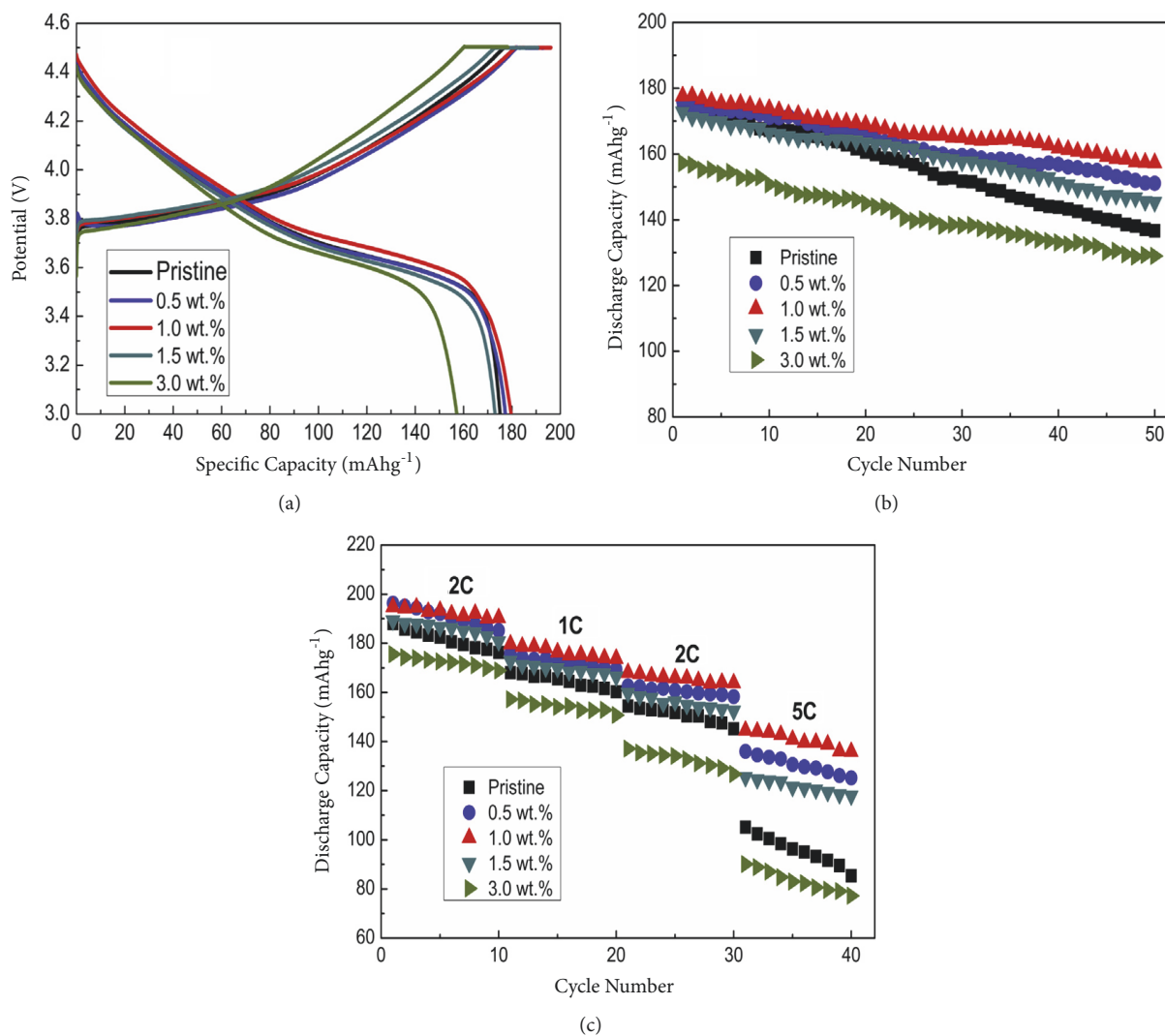


FIGURE 6: (a) Initial charge-discharge, (b) cycle performance, and (c) rate capability of TiO<sub>2</sub> coated NCM622 [33]. Copyright: Journal of Power Sources, 2014.

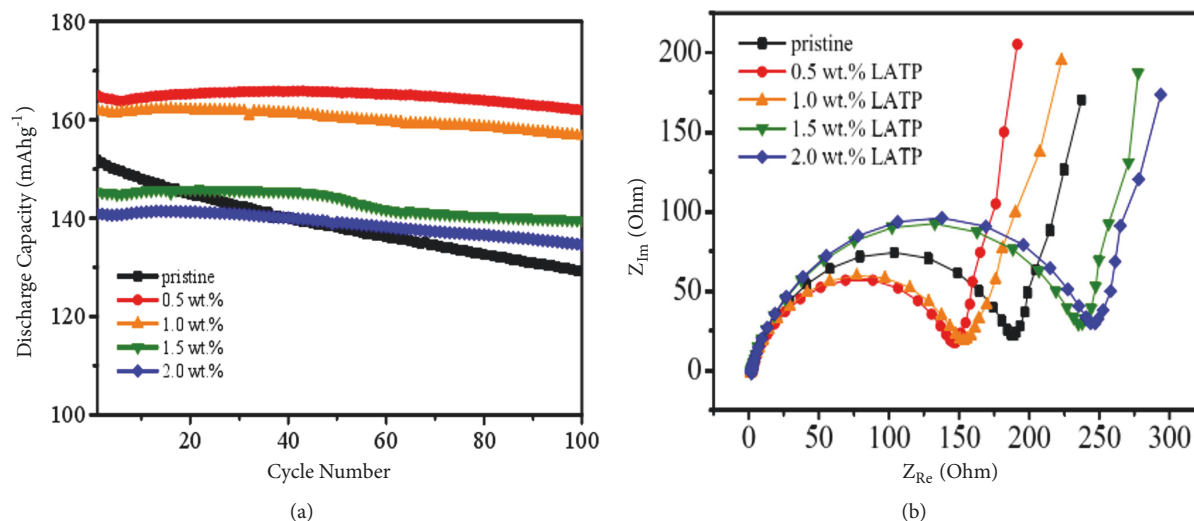
Chen et al. [33] prepared NCM622 cathode materials coated with 25-35nm TiO<sub>2</sub> by hydrothermal method and found that the improvement of the electrochemical performance for NCM622 should be attributed to the anatase TiO<sub>2</sub> coating. However, excess amount (3.0 wt.%) of inert TiO<sub>2</sub> coating reduced the discharge capacity and cyclic stability (Figure 6), because the thick anatase TiO<sub>2</sub> coating decreased the electrode-electrolyte interface area and suppressed the diffusion of Li-ion. At the same time, they also prepared Al<sub>2</sub>O<sub>3</sub> coated NCM622 and found that a layered structure of LiAlO<sub>2</sub> coating was formed, which decreased the contact area between the active material and electrolyte and reduced the interfacial resistance between the cathode and the electrolyte [34]. Li et al. [35] reported an effective surface modification for NCM622 that an Al-based MOF (NH<sub>2</sub>-MIL-53) was used as a precursor to produce MOF-derived alumina (MDA) coatings. The MDA layer showed well-dispersion and the amorphous nature and could not block the pathways of Li<sup>+</sup> ions. Moreover, the NCM622 cathode material was very

sensitive to the amount of ZrO<sub>2</sub> coating [36]. The ZrO<sub>2</sub> coating could enhance the stability of interface between the electrolyte and cathode material. However, the excess amount of ZrO<sub>2</sub> coating could negatively affect the discharge capacity and capacity retention owing to the decrease of electron conductivity in the thick ZrO<sub>2</sub> coating.

As is known, SiO<sub>2</sub> is corrosion resistant and has a poor conductivity. When the nano-SiO<sub>2</sub> was coated on the surface of NCM622 cathode materials, the thick SiO<sub>2</sub> coating could improve the cycle performance of NCM622 but reduce the conductivity at a high current density [37]. Meanwhile, silicate also has a high fusing point and hardness and may react with lithium hydroxide to form a layered structure of Li<sub>2</sub>SiO<sub>3</sub>, which rarely dissolves in acid or alkali environment [38]. For instance, Wang et al. [39] prepared the Li<sub>2</sub>SiO<sub>3</sub> coated NCM622 cathode materials by hydrolyzing the tetraethyl orthosilicate (TEOS) and found that the Li<sub>2</sub>SiO<sub>3</sub> coating improved the capacity retention, inhibited the corrosion reaction of the electrolyte, and enhanced the Li<sup>+</sup> diffusion. The

TABLE 3: Electrochemical properties of  $\text{Mn}_3(\text{PO}_4)_2$  coated and uncoated NCM622 cathode materials at different temperatures [41].

| Samples         | 25°C                         |                               | 60°C                         |                               |
|-----------------|------------------------------|-------------------------------|------------------------------|-------------------------------|
|                 | 1st cycle/ $\text{mAh}^{-1}$ | 50th cycle/ $\text{mAh}^{-1}$ | 1st cycle/ $\text{mAh}^{-1}$ | 50th cycle/ $\text{mAh}^{-1}$ |
| Pristine NCM622 | 165                          | 153                           | 175                          | 142                           |
| 0.5wt% NCM622   | 160                          | 149                           | 176                          | 160                           |

FIGURE 7: (a) Cycle life performance and (b) impedance curves of  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  coated NCM622 [43]. Copyright: Journal of Power Sources, 2016.

$\text{Li}_2\text{Si}_2\text{O}_5$  coated NCM622 cathode materials were prepared by sintering the lithium salt and  $\text{H}_2\text{SiO}_3$  [40]. The cyclic stability of modified NCM622 cathode material was better than that of unmodified sample. This indicates that the  $\text{Li}_2\text{Si}_2\text{O}_5$  coating could effectively reduce the residual lithium on the surface of cathode materials and prevent the corrosion of cathode materials from electrolyte.

The nanosized  $\text{Mn}_3(\text{PO}_4)_2$  coating was proposed to enhance the electrochemical and thermal properties of NCM622 [41]. As seen in Table 3, the discharge capacity and capacity retention of the  $\text{Mn}_3(\text{PO}_4)_2$  coated NCM622 were much higher than those of uncoated NCM622 at different temperatures. Furthermore, the thermal stability of  $\text{Mn}_3(\text{PO}_4)_2$  coated NCM622 was improved due to the strong  $\text{PO}_4$  covalent bonds. Because the  $\text{Li}_3\text{PO}_4$ , as a Li-ion conductor, was generated by  $\text{PO}_4^{3-}$  and  $\text{Li}^+$ , the increase of interfacial resistance for the  $\text{Mn}_3(\text{PO}_4)_2$  coated NCM622 was lower than that of the uncoated cathode material.

To improve the interface reaction and reduce the interfacial resistance, the Li-ion conductor  $\text{Li}_3\text{PO}_4$  is used for the modification of cathode materials. When the  $\text{Li}_3\text{PO}_4$  coated NCM622 was prepared with sol-gel method using citric acid as complexing agent [42], the  $\text{Li}_3\text{PO}_4$  coating could effectively hinder the phase transition (layered structure (R-3m)  $\rightarrow$  rock salt structure (Fm-3m)) of charged NCM cathode material at high temperature. The enhancement of thermal and electrochemical properties was mainly attributed to the  $\text{Li}_3\text{PO}_4$  coating, which improved the structural stability of the NCM622 material and suppressed the interface reaction between the electrolyte and cathode.

Some cathode materials with positive electrochemical properties are also used as coating materials. For example, the  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  coated NCM622 cathode materials were prepared with a sol-gel method and the effect of coating amount on the electrochemical properties of cathode materials was studied [43]. As shown in Figure 7, the cyclic stability of the coated cathode materials was improved. When the content of  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  coating was lower than 1.0 wt.%, the sample possessed higher discharge capacity and lower electrochemical impedance. If the amount of  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  coating was higher than 1.0 wt.%, the interfacial resistance of coated materials increased, and then the discharge capacity of  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  coated sample was lower than pristine. Actually, when the coating amount was more than suitable amount, the thick surface coating would be resistant to the movement of lithium ions. Similarly, the  $\text{Li}_{3x}\text{La}_{2/3-x}\text{TiO}_3$  coated NCM622 cathode material exhibited similar electrochemical performance to  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  coated NCM622 cathode material [44]. The dual conductive poly (3, 4-ethylenedioxythiophene)-co-poly (ethylene glycol) (PEDOT-co-PEG) copolymer was used as the coating material to improve the electrochemical performance of NCM622 [45]. Because the presence of a protective conducting polymer layer formed on the cathode also suppressed the growth of a resistive layer and inhibited the dissolution of transition metals from the active cathode materials, the cycling characteristics of copolymer coated NCM622 were more stable than those of the pristine. Lin et al. [46] found that the surface structural reconstruction (formation of a surface reduced layer, R-3m to Fm-3m transition) on

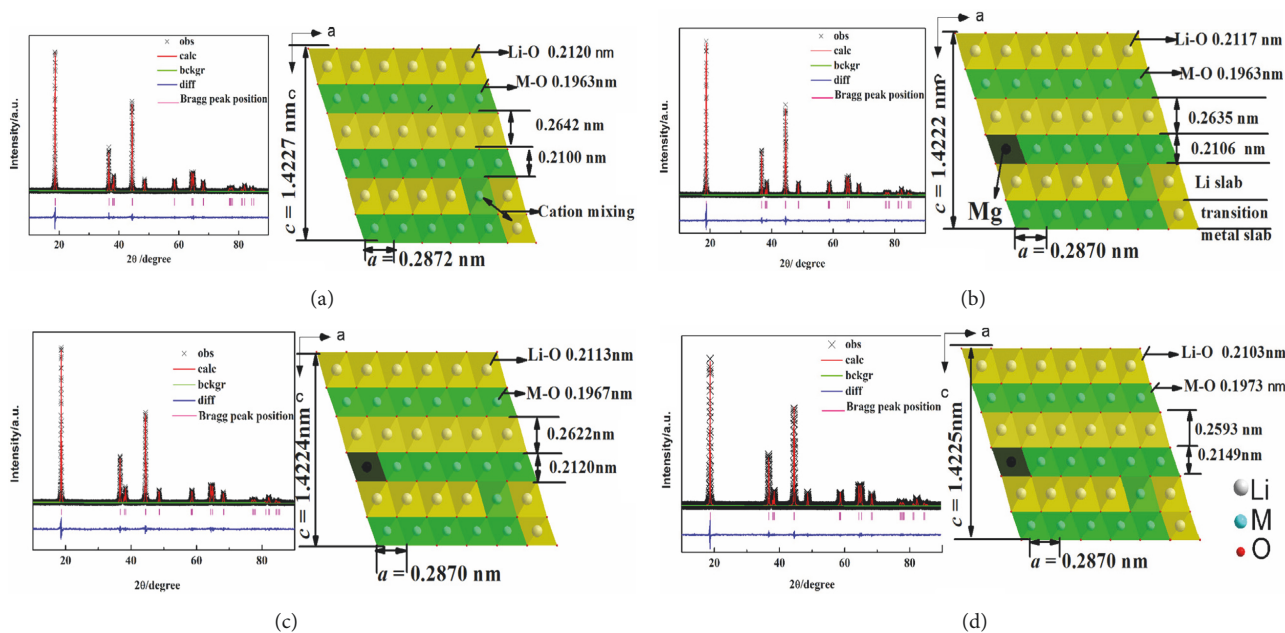


FIGURE 8: Rietveld refinement results for the prepared samples: (a) Mg-0, (b) Mg-1, (c) Mg-3, and (d) Mg-5 [48]. Copyright: Electrochimica Acta, 2015.

the surface of NCM<sub>xyz</sub> cathode materials exhibited a strong anisotropic characteristic, which predominantly occurred along lithium diffusion channels. This work sets a refined example for the study of surface coating modification.

**3.2. Doping Method of NCM622.** The electronic conductivity and the ionic conductivity of the NCM cathode material were increased via doping metal or nonmetal ions into the crystal lattice of the NCM cathode material. Meanwhile, the structural stability and thermal stability of the cathode materials were improved [47]. Different doping elements have different functions and properties, which may greatly influence the electrochemical performance of cathode materials.

For the NCM622 cathode materials, the doping elements should be aimed at improving the electronic conductivity and rate performance, such as Mg [49], Na [50], Nb [51], and Zr [52]. For example, Li(Ni<sub>0.6</sub>Co<sub>0.2</sub>Mn<sub>0.2</sub>)<sub>1-x</sub>Mg<sub>x</sub>O<sub>2</sub> cathode materials were prepared with coprecipitation method [49]. It was found that the doping of Mg element occupied the place of Co element. When the doping amount was 0.03, the electronic conductivity was nearly 100 times higher than that of the undoped NCM622. Similarly, Huang et al. [48] prepared Mg doped NCM622 cathode materials with coprecipitation and high temperature solid state reaction. As shown in Figure 8, the ratio of lattice parameters  $c$  and  $a$  ( $c/a$ ) increased after Mg doping. It was beneficial to improving the electrochemical performance of the battery. In addition, they also prepared Na-doped NCM622 cathode materials and found that the Na<sup>+</sup> doping reduced the mixing ratio of Li/Ni ions of the NCM622 cathode material [50]. As seen in Figure 9, the Li<sup>+</sup> diffusion coefficient of Na-doped NCM622 cathode materials was higher than that of

the pristine material, and the more the doping amount, the higher the Li<sup>+</sup> diffusion coefficient and the better the electrochemical properties. The results demonstrated that Na<sup>+</sup> doping could improve the chemical diffusion coefficients of Li<sup>+</sup> through enlarging the Li layer spacing. Similarly, the Li/Ni disorder degree was decreased, and the lithium slab spacing was enlarged through Mo doping, which was beneficial to facilitate the diffusion of lithium ions. Moreover, the Mo-modified material exhibited better rate performance and lower voltage fading in comparison to the unmodified material [53].

In fact, the battery will produce traces of HF during the circulation process of charge/discharge. If some doping elements could help to resist HF erosion, the electrochemical performance could be enhanced [51]. For the Nb-doped NCM622 material, the Li<sub>3</sub>NbO<sub>4</sub> thin film was formed at the grain boundaries and the surface of cathode material. The corrosion and decomposition of cathode materials were inhibited by Nb ions in the electrolyte. This indicated that the Nb doping was an effective method to improve the cathode's thermal stability. A particularly promising doping element for NCM is Zr, although its effect is still in dispute in the historical literature. As compared with the pristine cathode material, the Zr-doped LiNi<sub>0.56</sub>Zr<sub>0.04</sub>Co<sub>0.2</sub>Mn<sub>0.2</sub>O<sub>2</sub> enhanced the Li<sup>+</sup> conductivity and reduced the polarization phenomenon of battery during the charge-discharge cycles [52].

The electrochemical properties of cathode materials before and after modification with various materials were listed in Table 4. It is obvious that surface coating and element doping are the effective methods to improve the specific capacity, cycle life, and thermal stability of the cathode materials.



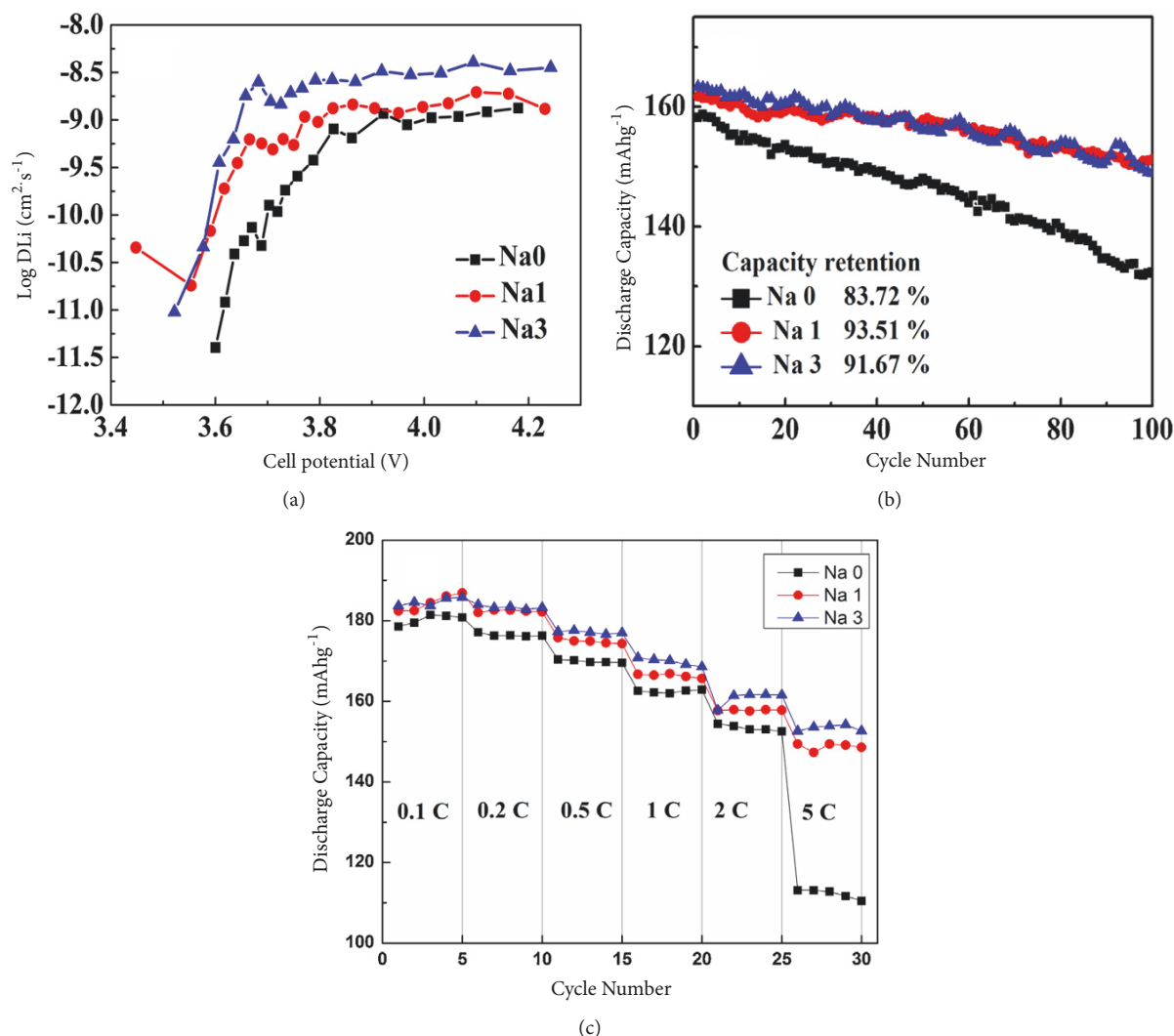


FIGURE 9: (a) Li<sup>+</sup> diffusion coefficient, (b) cyclic performance at 160mAhg<sup>-1</sup>, and (c) rate capability of Na<sup>+</sup> doped-NCM622 [50]. Copyright: Electrochimica Acta, 2016.

#### 4. Summary and Outlook

NCM622 cathode materials with the excellent electrochemical properties are promising in next generation lithium-ion batteries. Their cyclic stability and thermal stability have been significantly improved after surface coating or element doping. The various modification mechanisms for NCM622 mainly focus on the following aspects: The coating layer provides the isolation protection layer on the cathode material surface and greatly weakens the gap between the electrolyte and the electrode material. Meanwhile, the surface coating material enhances the ionic conductivity of the cathode material particles and shortens the Li<sup>+</sup> diffusion aisle during the charge-discharge cycles. The coating effectively reduces the electrochemical impedance and improves the battery cycle performance. The doping ions reduce the mixing of Ni<sup>2+</sup> and Li<sup>+</sup> in NCM622 cathode material and meliorate the aisle of Li<sup>+</sup> diffusion and then increase the stability of

the cathode material and impede the structural damage of NCM622 during the cycle of charging/discharging. Based on the structure and properties of cathode materials, the surface coating and ion doping are designed to improve their electrochemical and structural stability.

The research of Ni-rich cathode material (Ni content  $\geq$  60%) has been gradually paid more attention for the past few years. A high performance ternary material still faces many challenges related to structural instability and reaction mechanism at harsh temperatures. The future modification work should be carried out from the following aspects: Firstly, the energy density and safety of cathode material are supposed to be improved during the charge-discharge cycles. Secondly, the cyclic stability should be improved under the harsh environment conditions. Finally, in order to reduce the cost of production for commercial promotion, the preparation process of NCM622 should be optimized.

TABLE 4: Electrochemical performance of NCM622 before and after modification.

| Authors       | Modification methods   | Testing conditions | Pristine NCM622 (mAh/g)               | Modified NCM622 (mAh/g)                | Ref. |
|---------------|--|--------------------|---------------------------------------|--|------|
| Chen et al.   | 1wt% TiO <sub>2</sub> coating                                      | 1C, 2.8–4.5V       | 175.1(initial);<br>78.1%(50 cycles)   | 177.3(initial);<br>88.7%(50 cycles)    | [33] |
| Chen et al.   | 1wt% Al <sub>2</sub> O <sub>3</sub> coating                        | 1C, 3.0–4.5 V      | 176.8 (initial);<br>82.9%(30 cycles)  | 197 (initial);<br>91%(300 cycles)      | [34] |
| Li et al.     | 2.5wt% MDA coating   | 1C, 3.0–4.5 V      | 173.5(initial);<br>72.5%(100 cycles)  | 196.5(initial);<br>92.7%(100 cycles)   | [35] |
| Tao et al.    | 0.5wt% ZrO <sub>2</sub> coating                                    | 0.1C, 2.8–4.3V     | 133.7(initial);<br>83.8%(100 cycles)  | 146.6(initial);<br>83.8%(100 cycles)   | [36] |
| Cho et al.    | 1wt% SiO <sub>2</sub> coating                                      | 0.5C, 2.8–4.3V     | 168.1 (initial);<br>94%(50 cycles)    | 167.9 (initial);<br>95%(50 cycles)     | [37] |
| Fu et al.     | 2wt% Li <sub>2</sub> SiO <sub>3</sub> coating                      | 1C, 2.8–4.6 V      | 180(initial);<br>62.2%(200 cycles)    | 191.7 (initial);<br>73.4%(200 cycles)  | [38] |
| Wang et al.   | 3wt% Li <sub>2</sub> SiO <sub>3</sub> coating                      | 0.2C, 2.8–4.3V     | 196 (initial);<br>88.7%(100 cycles)   | 199 (initial);<br>85.2%(200 cycles)    | [39] |
| Liu et al.    | 1wt% Li <sub>2</sub> Si <sub>2</sub> O <sub>5</sub> coating        | 5C, 3.0–4.3 V      | 171.7 (initial);<br>81.6%(150 cycles) | 182.4 (initial);<br>86.4%(150 cycles)  | [40] |
| Cho et al.    | 0.5wt% Mn <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> coating     | 0.5C, 3.0–4.3V     | 153 (initial);<br>92.6%(50 cycles)    | 149 (initial);<br>93.3%(50 cycles)     | [41] |
| Choi et al.   | 1wt% Li <sub>3x</sub> La <sub>2/3-x</sub> TiO <sub>3</sub> coating | 0.5C, 3.0–4.3V     | 161.8 (initial);<br>56.6%(200 cycles) | 162.4 (initial);<br>87.2%(200 cycles)  | [43] |
| Ju et al.     | PEDOT-Co-PEG coating   | 0.5C, 2.8–4.3 V    | 10.7% capacity loss (100 cycles)      | 6.1% capacity loss (100 cycles)        | [45] |
| Fu et al.     | 3% Mg doping   | 5C, 3.0–4.3 V      | 126 (initial);<br>90.65%(30 cycles)   | 148 (initial);<br>95.81%(30 cycles)    | [49] |
| Huang et al.  | 1% Mg doping   | 1C, 2.8–4.3V       | 162.6(initial);<br>79.33%(100 cycles) | 169.7 (initial);<br>90.02%(100 cycles) | [48] |
| Huang et al.  | 1% Na doping   | 1C, 3.0–4.3 V      | 158.2(initial);<br>83.72%(100 cycles) | 162 (initial);<br>93.51%(100 cycles)   | [50] |
| Kaneda et al. | 3% Nb doping   | 2C, 3.0–4.1 V      | 150 (initial);<br>70.7%(500 cycles)   | 139 (initial);<br>91.4%(500 cycles)    | [53] |

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

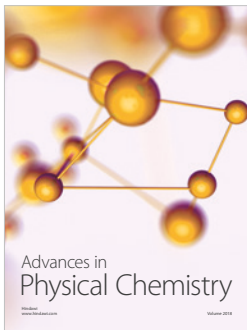
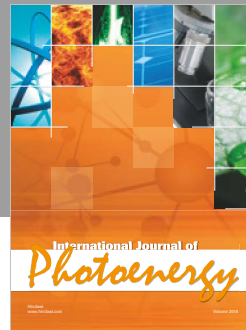
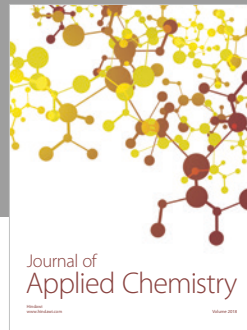
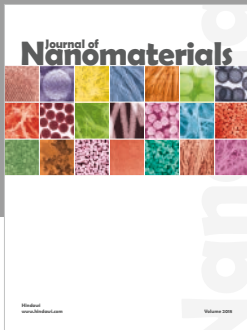
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