

# Design and Development of Cathode Materials for Rechargeable Batteries

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## 1. Introduction

Over the past two decades, rechargeable Li-ion batteries (LIBs) have been the de facto standard power source for electronic devices. However, as LIBs' applications are extended to electric vehicles and grid-scale energy storage systems, significant improvements in battery performance are required. In particular, since the cathode accounts for the largest portion in LIBs in terms of weight and cost [1], it is commonly assumed that the cathode chemistry primarily determines the energy density and cost of the cell.  $\text{LiCoO}_2$  (LCO) and  $\text{Li}[\text{Ni}_a\text{Co}_b\text{Mn}_c]\text{O}_2$  ( $a + b + c = 1$ ; NCM) are currently used as standard cathode materials in commercial LIBs for small electronic devices and electric vehicles, respectively. It is important to consider that small variations in material composition have a considerable impact on the operating voltage of cathodes [2], which has led many scientists to believe that the key to improving energy density lies in the optimization of cathode chemistry. Furthermore, cathode materials such as LCO and NCM face price volatility problems due to the supply instability of cobalt and nickel elements [3]. In this context, formidable research efforts are constantly being made to optimize the performance of cathode materials or to develop a new concept of cathode materials.

This Special Issue, entitled "Cathode Materials for Rechargeable Batteries", aims to present recent breakthroughs in cathode material design, discovery, and engineering. Topics of interest for publication include, but are not limited to:

- Discovery of new cathode materials;
- Chemical/structural engineering of cathode materials;
- Degradation mechanism analysis;
- Synthesis of cathode materials;
- Electrode design;
- Development of high-nickel layered oxides electrodes;
- Development of lithium-rich layered oxides electrodes;
- Development of cation-disordered rocksalt transition metal oxides electrodes;
- Development of next-generation Li-ion batteries such as Li-air, Li-sulfur, Li-organic batteries;
- Cathode materials for sodium-ion batteries.

## 2. Design and Development of Cathode Materials for Li-Ion Batteries

Because range anxiety is one of the key worries of EV consumers, the energy density of cathode material is becoming increasingly important [4]. An intuitive way to improve the specific energy density of an NCM electrode is to increase the content of nickel, which serves as a redox center during electrochemical cycling. On that account, NCM series has been developed in the direction of increasing the nickel amount (NCM-111  $\rightarrow$  NCM-523  $\rightarrow$  NCM-622  $\rightarrow$  NCM-811) [1]. Still, numerous research teams are working to fully commercialize NCM-811 and produce high-nickel layered oxide electrodes with far greater nickel content than commercial NCM. Unfortunately, an increase in nickel content has been known to hasten structural degradation of electrodes by enhancing cation mixing,



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surface reconstruction, strain, lattice mismatch, and microcrack generation, all of which inevitably compromise capacity retention [5]. To improve the structural stability of high-nickel layered oxide electrodes, various engineering strategies such as doping, surface coating, and microstructural engineering have been developed [6]. On the other hand, Li-rich layered oxides electrodes have also received much attention as promising candidates of high-energy-density cathode materials [7]. Li-rich layered oxides refer to a class of materials in which a portion of the TM sites in conventional layered-type cathodes are substituted with Li ions. It is widely accepted that the utilization of oxygen redox during the charging and discharging process enables the extraction of a high capacity from Li-rich layered oxides [7]. However, Li-rich layered oxides electrodes suffer from severe voltage fade and hysteresis problems, which have been linked to electrode structural degradation [8]. Interestingly, recent research has identified intrinsic links between the oxygen redox mechanism and the structural degradation mechanism [9], and various engineering strategies have been developed to address voltage depression problems based on these such understandings. In addition to electrodes with a layered-type structure, some research groups have attempted to develop cationic disordered rocksalt oxide cathodes (DRX) for which there is no need to restrict the type of transition metals used to maintain the layered structure [10]. Although typical Li-stoichiometric DRX electrodes have known to show poor rate capability, the pioneering work of Lee et al. revealed that moderate rate capability can be achieved with Li-rich DRX because an abundance of Li ions produces a well-connected Li percolation network [11]. Nevertheless, the practical implement of DRX cathodes requires further improvement in terms of energy density, rate capability, and cycling stability. If the energy storage scale increases by several orders of magnitude in the coming decades, the production of cathodes is expected to face a supply crisis related with transition metals [3]. In this regard, long-term efforts are being made to develop next-generation batteries using transition metal-free cathodes such as Li-air batteries and Li-organic batteries [12,13]. Although these next-generation batteries can provide significant economical and environmental benefits, their energy efficiency is remarkably low at the current stage, requiring significant improvements in materials' properties.

### 3. Conclusions

This Special Issue, entitled “Cathode Materials for Rechargeable Batteries”, are inviting original research articles and reviews on the synthesis, characterization, fabrication, engineering, and applications of cathode materials for rechargeable batteries. We firmly believe that the findings of this Special Issue will greatly contribute to the battery community, expanding our understanding of cathode materials.

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