Sustainable Recycling Technologies of Lithium-Ion Battery Cathode: Recent Advances and Future Perspectives

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Abstract. With the rapid development of electric vehicles and energy storage systems, the amount of waste lithium-ion batteries has increased sharply, and the demand for recycling has become increasingly urgent. This paper focuses on the green recycling technology of lithium-ion battery cathode materials, aiming to achieve resource recycling and environmental sustainability. This review analyzes the characteristics of cathode materials such as $LiCoO_2$, NMC, $LiFePO_4$ and $LiMn_2O_4$ and their influence on the applicability of recycling methods. The principles, efficiency and scale-up potential of green recycling technologies, such as solid-state sintering, hydrothermal method, molten salt treatment and electrochemical method, are discussed in detail. These technologies recover key metals such as lithium, cobalt and nickel in a low-impact manner while reducing greenhouse gas emissions and secondary pollution. It was shown that green recycling technology has advantages in material purity and energy efficiency, but energy consumption and cost need to be optimized to achieve scale. In the future, through technological innovation and policy support, lithium-ion battery recycling can help the development of the circular economy and provide guarantees for sustainable energy.

Keywords: Lithium-ion battery recycling; green recycling technologies; cathode materials; circular economy.

1. Introduction

The swift expansion of electric vehicles (EVs) along with developments in consumer electronics and energy storage systems has resulted in both a massive increase in lithium-ion batteries (LIBs) usage and a significant rise in discarded batteries. Battery recycling serves as essential practice for environmental sustainability while also providing vital opportunities for both resource recycling and economic advantages. Through worldwide regulatory measures, multiple frameworks have encouraged the advancement of battery recycling practices [1]. According to the EU 2023 Battery Regulation, battery producers must reach a 70 % recycling rate for LIBs by 2030 and finance recycling projects through EPR to support green technology development. Moreover, the battery recycling urgency becomes apparent when examining market trends, including waste battery expansion alongside cost changes for metals like lithium and cobalt and resource limitations. The global waste LIBs are predicted to hit millions of tons by 2030 and recycling becomes economically viable based on the price changes of lithium and nickel metals. For example, direct recycling of $LiFePO_4$ can reduce production costs by 20 % compared to synthesizing new cathodes. However, traditional battery processing techniques, including pyrometallurgy and hydrometallurgy, fail to support sustainable development requirements because they consume excessive energy and create pollution while wasting resources. Traditional battery treatment methods mainly include pyrometallurgy and hydrometallurgy [2]. Pyrometallurgy recovers metals through high-temperature smelting, but it has high energy consumption, emits a large amount of greenhouse gases, and produces secondary pollutants such as waste slag. Hydrometallurgy (acid leaching) uses acidic solutions to extract metals. Although it is more selective than pyrometallurgy, it is difficult to treat acidic waste liquids, and the process is complex and costly. In addition, traditional methods generally face challenges such as low efficiency, limited recovery rate, and secondary pollution of the environment [3]. These limitations have promoted the development of green recycling technology to achieve higher resource recovery rates and lower environmental impacts.

Green recycling technologies leverage chemical and physical principles to regenerate cathode materials with minimal ecological footprint. Current research focuses intensely on green recycling technologies development, including solid-state sintering, hydrothermal method, molten salt treatment and electrochemical method [4]. Solid-state sintering restores crystal structures through high-temperature diffusion, while the hydrothermal method uses aqueous solutions for selective metal extraction. Molten salt processing employs molten salts as reaction media to facilitate metal oxide formation, and electrochemical methods utilize electric fields for precise metal recovery. These approaches contrast with pyrometallurgy by avoiding high-temperature combustion and reducing greenhouse gas emissions. In addition, emerging technologies like bioleaching and microwave-assisted recycling reduce greenhouse gas emissions by 50-70 %, aligning with global carbon neutrality goals. Direct recycling minimizes waste generation, as it avoids the production of secondary pollutants like slag. Additionally, recovering critical metals (e.g., cobalt, nickel) reduces the need for mining, which is associated with deforestation and water pollution.

This review explores four key green recycling technologies for LIB cathode materials: solid-state sintering, hydrothermal method, molten salt processing, and electrochemical methods. These methods aim to recover valuable metals (e.g., lithium, cobalt, nickel, manganese) and restore cathode performance while minimizing environmental impact. Firstly, the characteristics of different types of cathode materials (e.g., $LiCoO_2$, NMC, $LiFePO_4$, $LiMn_2O_4$) with distinct characteristics was analyzed, which may affect the applicability of the recovery method. Then the principles, methods and applicability of green recycling technologies were described respectively. Specific case studies from recent literature are presented, highlighting the recovery of cathode materials and their performance in subsequent testing. The focus is on the mechanisms, efficiency, scalability, and ongoing research to address challenges like energy consumption and material purity. Finally, future trends and challenges in LIB cathode material recycling were discussed.

2. Characteristics of Lithium-Ion Battery Cathode Materials

Cathode materials are key to the performance of LIBs, determining energy density, cycle life, safety, and cost. Their electrochemical performance depends on chemical composition and crystal structure, and different characteristics affect the selection of cathode materials in specific applications and the suitability of green recycling processes [5, 6]. The main types include lithium cobalt oxide ($LiCoO_2$), nickel manganese cobalt oxide (NMC), lithium iron phosphate ($LiFePO_4$), and lithium manganese oxide ($LiMn_2O_4$).

$LiCoO_2$ has a layered oxide structure (R-3m space group), with lithium ions embedded in the cobalt oxide interlayer, and has high voltage and high capacity characteristics. It has an energy density of about 150-190 Wh/kg and a stable discharge voltage (about 3.9 V), and is widely used in consumer electronics. However, due to structural instability, its capacity decays after about 500 cycles. In addition, the reliance on cobalt increases battery costs and raises environmental issues due to the toxicity and scarcity of cobalt.

NMC cathodes combine nickel, manganese and cobalt in different proportions (e.g. $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ or NMC811), where nickel improves capacity, manganese enhances structural stability and cobalt improves conductivity. NMC cathodes (e.g. NMC111, NMC532, NMC811) strike a balance between capacity (160-200 Wh/kg), structural stability and cost-effectiveness and are widely used in EVs. High-nickel NMC811 has a capacity of up to 200 mAh/g and a cycle life of about 1,000 cycles after optimized formulation, but high-nickel variants face the risk of thermal runaway.

$LiFePO_4$ adopts an olivine structure (Pnma space group), and iron and phosphate groups provide thermal stability and safety, although its low electronic conductivity requires carbon coating to improve performance. $LiFePO_4$ is favored for its excellent thermal stability, ultra-long cycle life (>2,000 times) and low cost. It has a low operating voltage (about 3.4 V) but excellent safety, making it suitable for stationary energy storage. However, its low energy density (about 120-160 Wh/kg) limits its use in high-energy applications.

$LiMn_2O_4$ has a spinel structure (Fd-3m space group), a capacity of about 120 mAh/g, good safety, moderate cost, and a high voltage (about 4.0 V). However, due to the dissolution of manganese in the electrolyte and Jahn-Teller distortion at high temperatures, its capacity decays and its cycle life is about 500-800 times.

3. Green Recycling Technologies for Lithium-Ion Battery Cathode Materials

3.1. Solid-State Sintering

Solid-state sintering is a green recycling method that achieves low environmental impact regeneration by restoring the crystal structure of spent LIB cathode materials. The process involves collecting spent cathodes (such as $LiCoO_2$, NMC), grinding them into fine powders, mixing them with lithium additives (such as Li_2CO_3, LiOH), and sintering them at 600-900 ℃ in an argon atmosphere for 6-12 hours to prevent oxidation. Material characterization is performed by X-ray diffraction (XRD) and scanning electron microscopy (SEM), the crystal structure recovery is evaluated [7], and the efficiency is evaluated by electrochemical testing, which can produce cathode materials with performance comparable to that of the original material (about 140 mAh/g). This method avoids the generation of waste liquid, reduces secondary pollution, and has a simple process.

For example, Schwich et al. recovered solid-state sintered $LiCoO_2$ from discarded laptop batteries [8]. They mixed the ground cathode material with Li_2CO_3 and sintered it at 800°C for 10 hours to successfully regenerate $LiCoO_2$ with a layered structure, which was verified by XRD. The recycled material was used to make button batteries with a capacity of 138 mAh/g, which is about 95 % of the original material. This method effectively repairs structural defects, but the energy consumption is high (250 kWh/kg). Doping with 2 % Al can improve the cycle stability by 15 %, but the energy consumption problem still needs to be solved.

Solid-state sintering has a significant effect on the regeneration of $LiCoO_2$ and NMC, but the high operating temperature (600-900 °C) leads to high energy consumption. High energy consumption and sensitivity to temperature control limit its large-scale application. Researchers are exploring the use of catalysts (such as Li_3PO_4) for low-temperature sintering (400-500 °C), which can reduce energy consumption by 25 % while maintaining 90 % capacity retention. In addition, integrating renewable energy such as solar energy into the sintering process can further improve sustainability.

3.2. Hydrothermal Method

The wet heat method uses aqueous solutions at high temperature (150-250 °C) and high pressure to extract and regenerate cathode materials. It is suitable for complex cathodes such as NMC and can recover lithium and transition metals (nickel, manganese, cobalt) with high purity. The process involves dissolving the spent cathode in a water-based medium, followed by precipitation or recrystallization. The spent cathode is immersed in deionized water containing 1 M citric acid and reacted in a stainless steel autoclave at 150-250 °C and 2-5 MPa pressure for 6-24 hours. The solution promotes the dissolution of lithium and transition metals, which are then precipitated with NaOH to form metal hydroxides. The recovery rate of NMC cathodes is generally over 95 %, and nickel, manganese, and cobalt are precipitated in the form of hydroxides. Purity is optimized by adjusting pH (6-8) and temperature.

Yu et al. used the wet heat method to recover NMC532 from electric vehicle batteries [9]. The cathode was treated in a citric acid solution at 200 °C for 12 hours, and the recovered lithium was in the form of Li_2CO_3, and the transition metals were precipitated as hydroxides with a purity of 96 %. The regenerated NMC532 was assembled into a soft-pack battery with a discharge capacity of 155 mAh/g, which is about 90 % of the original material. This method avoids the use of toxic solvents and conforms to the principles of green chemistry. However, the long reaction time (6-24 hours) and the need for a dedicated autoclave increase the cost. Optimizing the conditions of 200 °C and pH 7 can shorten the reaction time to 8 hours, improving the scale-up potential.

The long reaction time (6-24 hours) and reliance on autoclaves of the wet heat method limit industrial scale-up. The microwave-assisted wet heat process, which was run at 200 °C for 2 hours, showed the potential to shorten the reaction time by 80 % and achieve a metal recovery rate of 94 %. In the future, continuous flow wet heat systems should be optimized to shorten the processing time and improve the scale-up capability.

3.3. Molten Salt Processing

Molten salt treatment uses low melting point salts (such as NaCl, KCl) as reaction media to recover cathode materials at medium temperature (300-600 °C), promote lithium extraction and metal oxide formation, and generate recyclable precursors. For example, a eutectic mixture of NaCl and KCl (molar ratio 1:1) is heated to 300-600 °C and added to the waste NMC cathode. Molten salt is used as a solvent to extract lithium as Li_2CO_3, and metal oxides are formed after 4-8 hours. The salt is recovered by filtration and reused.

Liu et al. used NaCl-KCl melt at 500 °C for 6 hours to recover NMC111 in electric vehicle batteries [10]. The recovery efficiency of lithium carbonate was 92 %, and oxides of nickel, manganese, and cobalt were obtained, which were used to synthesize new NMC111 cathodes. In full battery tests, the recycled material capacity was 145 mAh/g, and 93 % of the performance of the original material was retained after 100 cycles. Compared with pyrometallurgy, this technology consumes less energy and produces less waste. The 93 % capacity retention of recycled NMC111 indicates its suitability for reuse in electric vehicle batteries. However, the purification and handling of salts pose challenges for industrial applications. The researchers solved the salt purification problem by adding 1 % $CaCl_2$ as a flux and explored a reusable salt system to improve sustainability.

3.4. Electrochemical Methods

Electrochemical methods use electric fields to selectively extract lithium and metals from spent cathodes with high precision and low environmental impact. The process involves applying a potential in an electrolyte to dissolve the cathode material, followed by metal electrodeposition. Using a three-electrode system (working electrode: spent cathode, reference electrode: Ag/AgCl, counter electrode: Pt), 1 M LiCl was used as the electrolyte, a 2-3 V potential was applied for 10-15 hours to dissolve the metal, followed by electrodeposition at -1.5 V.

For example, Yu et al. conducted electrochemical recovery experiments on $LiCoO_2$ from smartphone batteries [11]. Using 1 M LiCl electrolyte, a 2.5 V potential was applied for 12 hours, and the lithium recovery rate reached 96 %. The recovered cobalt was electrodeposited in the form of $Co(OH)_2$ and subsequently converted to $LiCoO_2$. The regenerated $LiCoO_2$ was tested in a button cell with a capacity of 135 mAh/g, which is about 94% of the original material. The recovery efficiency of lithium from $LiCoO_2$ can exceed 95% using aqueous electrolytes. Advantages include high selectivity and no need for highly corrosive chemicals.

However, low processing speeds and high equipment costs limit scalability. Optimizing electrode materials and electrolyte composition can improve efficiency. For example, using optimized electrodes (such as carbon felt) can increase processing speed by 20 %. 3D printing electrodes and alternative electrolytes (such as ionic liquids) can reduce costs by 40 % while maintaining 95 % recycling efficiency. These advances can make electrochemical recycling applicable to low-cost materials such as $LiMn_2O_4$, expanding its application in power tools and grid energy storage.

3.5. Emerging Recycling Technologies

3.5.1 Bioleaching and biotechnological methods

Bioleaching uses microorganisms (such as Acidithiobacillus ferrooxidans and Leptospirillum ferriphilum) to extract metals from spent cathodes through a biooxidation process. The method operates at ambient temperature (20-40 °C) and significantly reduces energy consumption compared to pyrometallurgical methods. For example, bioleaching could recover 90 % of cobalt and 85 % of lithium from spent $LiCoO_2$ cathodes using sulfur-based media within 10 days [12]. The resulting products—metal sulfates—can be precipitated as hydroxides for battery manufacturing. However, long processing times and sensitivity to pH conditions (optimal 1.5-2.5) limit scalability. Future research aims to genetically engineer bacteria to improve metal extraction efficiency and shorten processing time.

3.5.2 Microwave-assisted recycling

Microwave-assisted recycling uses microwave radiation to quickly and uniformly heat cathode materials, shortening processing time and reducing energy consumption. For example, 95 % of lithium carbonate and metal oxides can be recovered by treating spent NMC cathodes with a reducing agent such as carbon black under 500 W microwaves for 30 minutes [13]. This method is particularly effective for high-nickel NMC variants because it reduces the risk of thermal runaway caused by traditional heating. Current research focuses on optimizing microwave frequency and power to further improve energy efficiency and material purity.

3.5.2 Direct recycling and relithiation

Direct recycling aims to restore spent cathode materials to their original state without decomposing them into elemental components. Relithiation is a subset of direct recycling, which replenishes lithium in degraded cathodes by chemical or electrochemical methods. Recent studies have successfully relithiated spent $LiFePO_4$ cathodes by immersing them in lithium iodide solution at 80 °C for 24 hours, with a capacity of 145 mAh/g, which is comparable to that of the original material [14]. This method is suitable for stable $LiFePO_4$ and can extend the life cycle of stationary energy storage batteries. However, direct recycling is less effective for materials with severe structural degradation such as high-nickel NMC. Innovations in automated sorting and relithiation technologies are expected to improve their applicability.

4. Conclusion

With the surge in demand for LIBs for EVs, renewable energy storage, and portable electronics, the sustainable recycling of cathode materials has become increasingly important. Green recycling technologies offer promising solutions for the sustainable recycling of LIB cathodes, among which solid-state sintering excels in material recovery, hydrothermal methods lead in purity, molten salt treatment dominates in energy efficiency, and electrochemical methods stand out in precision. Although scale-up remains challenging, continued innovation in process optimization and materials science is expected to address these limitations and support the development of a circular economy for LIBs.

Major challenges include high energy consumption for sintering, long reaction time for hydrothermal methods, salt management issues for molten salt treatment, and high equipment costs for electrochemical methods. Future research may consider adopting emerging technologies such as bioleaching, microwave-assisted recycling, and direct recycling, while improving existing methods. For example, developing low-temperature sintering catalysts (e.g., Li_3PO_4); Reducing the hydrothermal reaction time by microwave assistance; Improving the recyclability of salts using ionic liquid additives; Reducing the cost of electrochemical devices using 3D printed electrodes.

These innovations are expected to bring significant environmental benefits (such as reduced emissions and waste) as well as economic advantages (including cost savings and resource security). Supportive policies and industry collaboration will be key to overcoming scale and cost barriers, ensuring that the LIB industry achieves a fully circular economy by 2030.

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