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Research Progress on Green Recycling and Regeneration Technology of Waste Lithium Battery Cathode Materials

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Abstract

The large-scale development of new energy vehicles and energy storage industry has driven the continuous growth of demand for lithium batteries, and waste lithium batteries have entered the stage of centralized retirement. According to the forecast of China Commerce Industry Research Institute in 2024, the retired volume of power batteries in China will reach 1.04 million tons by 2025. The cathode materials of waste lithium batteries contain valuable metals such as lithium, cobalt, and nickel. Disorderly disposal not only causes resource loss, but also soil and air pollution due to heavy metal leakage and electrolyte volatilization. Green recycling and regeneration technology is the core path to achieve the coordinated development of resource recycling and ecological environmental protection. Currently, the industry has formed three mainstream technology systems: pyrometallurgical, hydrometallurgical, and direct regeneration. After technical optimization, The lithium recovery rate of low-temperature pyrometallurgical process exceeds 60%; The green hydrometallurgical process has been industrialized and verified, with a lithium recovery rate of 96.5% and a comprehensive recovery rate of 99.6% for cobalt, nickel, and manganese; Direct regeneration technology can achieve crystal structure repair of cathode materials, and the performance of regenerated products approaches that of native materials. This article combines the characteristics of cathode materials and the urgency of recycling, summarizes the research progress of three major green technologies, analyzes the optimization results of processes, reagents, and equipment, explores the bottlenecks of industry technology and industrial development, looks forward to the trend of low-carbon, high-value, and fully closed-loop development, and provides reference for the research and application of related technologies.

Keywords

Waste Lithium Batteries; Cathode Materials; Green Recycling; Regeneration Technology; Resource Utilization.

1. Introduction

The rapid development of the new energy industry has promoted the widespread application of lithium batteries in the fields of new energy vehicles, portable electronic devices, and energy storage. According to statistics from the Ministry of Industry and Information Technology in 2024, the number of new energy vehicles in China has exceeded 31.4 million, and power batteries are entering a wave of large-scale retirement. The cumulative retirement volume is expected to exceed 3.5 million tons by 2030. China's key raw materials such as lithium, cobalt, and nickel have a high degree of external dependence, and the mining of primary minerals causes significant damage to the ecological environment. Recycling and regeneration are important ways to ensure the independent and controllable nature of the industrial chain, reduce external dependence on resources and mitigate environmental burdens. A large number of retired lithium batteries that have not been properly disposed of are prone to environmental problems such as heavy metal pollution and electrolyte leakage, exacerbating ecological

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governance pressure. Green recycling is also a key link in the sustainable development of the industry. The main cathode materials for waste lithium batteries are lithium cobalt oxide (LCO), nickel cobalt manganese ternary (NCM), and lithium iron phosphate (LFP), with a valuable metal content of up to 20%–30%. They are known as "urban mines" and have extremely high recycling value [1]. The traditional recycling process has drawbacks such as high energy consumption, low metal recovery rate, and serious secondary pollution. The lithium recovery rate of the traditional pyrometallurgical process is less than 50%, while the hydrometallurgical process is prone to producing a large amount of acidic wastewater, which contradicts China's green and low-carbon development concept. In 2024, the Ministry of Industry and Information Technology revised the "Industry Standard Conditions for Recycling and Utilization of New Energy Vehicle Power Batteries", raising the standard for lithium smelting recovery rate to 90% to force recycling technology to upgrade towards green and efficient. This article focuses on the green recycling and regeneration technology of waste lithium battery cathode materials, summarizes the progress and key technological breakthroughs in mainstream green processes, analyzes the existing challenges and development trends in the industry, and provides theoretical and practical support for the standardization and low-carbon development of the industry.

2. Characteristics of Cathode Materials for Waste Lithium Batteries and the Urgency of Recycling

The composition and structure of waste lithium battery cathode materials directly affect their recycling value and difficulty. Currently, the mainstream cathode materials on the market are mainly divided into three categories: lithium cobalt oxide (LCO), which is mostly used in portable electronic devices such as mobile phones and tablets, with cobalt content of up to 15%–20% and lithium content of about 1%–2%; Nickel cobalt manganese ternary (NCM) materials are widely used in new energy vehicle power batteries, with nickel content of 12%–20% and cobalt content of 5%–15%, making them the most valuable resource among the three types of materials; Lithium iron phosphate (LFP) is mainly suitable for energy storage equipment and low-end electric vehicles, rich in iron, lithium, and phosphorus elements, and has relatively lower recycling difficulty [2]. Lithium, cobalt and other metals are already scarce mineral resources. The mining of primary minerals not only takes several years or even more than ten years, but also causes serious ecological problems such as vegetation damage and soil desertification. On the other hand, the metal content of cathode materials in retired batteries is much higher than that of primary minerals. The cobalt content in ternary materials alone is equivalent to 10–15 times that of primary cobalt minerals. The economic and environmental benefits of recycling and utilization are prominent. According to GGII and the China Industrial Research Institute, the retirement of power batteries in China is expected to reach 1.04 million tons by 2025, corresponding to a value of over 100 billion RMB in recyclable metal resources. However, the current standardized recycling rate in the industry is still less than 25%, and a large number of waste batteries flow into informal recycling channels. These batteries, after being dismantled and burned in small workshops, not only cause unnecessary waste of valuable resources, but also allow heavy metals to infiltrate the soil and pollute water sources. Fluoride and organic solvents in electrolytes can also release toxic gases, which not only damage the ecological environment but also threaten human health. From a policy perspective, China has established a power battery traceability management platform, actively promoted the extended producer responsibility system, clarified and standardized the technical indicators of recycling enterprises, required the electrode powder recovery rate to be no less than 98%, and steadily promoted the industry's transformation from extensive processing to green and efficient [3]. It is thus evident that the recycling and utilization of waste lithium battery cathode materials is

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not only an important measure to ensure China's resource security, but also an inevitable requirement to practice the concept of green development. Its urgency and practical significance are self-evident.

3. Research Progress on the Greening of Pyrometallurgical Recycling Technology

Pyrometallurgical recycling is the earliest technological path to achieve industrialization in the field of waste lithium battery recycling. The core is to use high-temperature calcination to decompose the organic matter and binder in the cathode materials, achieve the enrichment of valuable metals, and then obtain metal elements or compounds through subsequent purification processes. The traditional pyrometallurgical process requires operation at a high temperature of 1300–1500 °C, which not only leads to high energy consumption but also consumes 800–1200 kWh per ton of waste batteries. The lithium recovery rate is only maintained at a low level of 40%–50%. During the roasting process, toxic waste gases such as dioxins are also easily generated, putting great pressure on environmental protection and management. This process still has shortcomings such as severe equipment corrosion, complex metal separation and purification processes, and high processing costs, which are difficult to adapt to the large-scale application of small and medium-sized enterprises and contradict the industry's low-carbon and efficient development direction. Against the backdrop of advancing the dual carbon strategy and increasingly stringent industry environmental standards, the green upgrade of pyrometallurgical recycling has become the core direction of industry research. The research focuses on low-temperature transformation, process synergy optimization, and flue gas resource utilization, in order to significantly reduce energy consumption and environmental load. Low-temperature roasting technology lowers the roasting temperature to 600–800 °C by introducing efficient reducing agents and fluxes, reducing energy consumption by about 40% compared to traditional processes, and synchronously increasing lithium recovery rate to 60%–70% [4]. The "layered roasting collaborative leaching" process developed by the team from Jiangxi University of Science and Technology relies on the sulfur vapor released from pyrite to break down the crystal structure of lithium cobalt oxide, which not only lowers the roasting temperature but also reduces the amount of aqueous leaching residue by 66.71%, greatly simplifying the subsequent purification process. The technology of flue gas resource utilization practices the idea of "Treatment of Waste with Waste", by introducing carbon dioxide from the roasting tail gas into the leaching solution to complete carbonization and lithium extraction. The lithium selective leaching rate exceeds 97%, and the cost of lithium extraction is also reduced by 20%. At present, the green pyrometallurgical process has highlighted its advantages in the pretreatment of complex component retired batteries [5], and is gradually evolving towards a combined process of "low-temperature calcination–wet purification", achieving a balance between treatment efficiency and metal recovery rate, becoming the preferred technical solution for small and medium-sized recycling enterprises.

4. Research Progress on Green Wet Recycling Technology

The wet recycling process, relying on the core advantage of high recovery rate of valuable metals and high purity of products, has become the leading technology for the industrialization of resource recycling of waste lithium battery cathode materials [6]. Its core processes include acid leaching, extraction, and precipitation, and rely on chemical reaction mechanisms to achieve selective separation and high-purity purification of valuable metals. The traditional hydrometallurgical process mostly uses sulfuric acid–hydrogen peroxide mixture as the core reaction system, and the metal recovery rate can be maintained in the range of 92%–95%.

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However, there are shortcomings such as high consumption of leaching reagents, large output of acidic wastewater (3–5 tons of acidic waste liquid accompany a single ton of waste batteries), high equipment corrosion rate, and high operation and maintenance costs for subsequent waste liquid treatment. In recent years, the green iteration research of wet recycling technology has focused on the replacement application of environmentally friendly leaching agents and the simplification and optimization of process flow, aiming to achieve the goal of resource recycling with low environmental load and high selectivity.

The research and industrial application of green leaching agents have achieved substantial innovation, gradually completing the iterative replacement of traditional strong acid reaction systems. The ternary deep eutectic solvent of choline chloride ascorbic acid ethylene glycol can achieve leaching efficiencies of 96% and 98% for lithium and iron, respectively, under leaching conditions of 80 °C and 1 h. The system has no toxic reagent residues and the solvent can achieve closed-loop reuse, significantly reducing environmental pollution hazards; The citric acid vitamin C complex leaching system can efficiently dissolve the cathode materials at room temperature, avoiding the corrosion of reaction equipment by high-temperature strong acids. The wastewater output is reduced by more than 30% compared to traditional processes. The iterative upgrade of directional separation technology further enhances product purity. In the 2024 technical white paper, Bangpu Cycle disclosed that its independently developed DRT directional separation technology can achieve a comprehensive recovery rate of 99.6% for nickel, cobalt, and manganese, and a lithium recovery rate of 93.8%. The purity of the regenerated metal meets the battery grade high-purity standard (99.9%) [7]. At present, leading companies in the industry such as Greenmei and Bangpu Recycling have established 10,000-ton green hydrometallurgical recycling production lines, accounting for over 60% of the industry's production capacity. They rely on refined technologies such as membrane separation and evaporation crystallization to dispose of waste liquids, achieve closed-loop water resource reuse, and further strengthen the green attributes and economic efficiency of the process.

5. Research Progress on Direct Regeneration Technology

Direct regeneration technology is different from the "disassembly extraction" mode of pyrometallurgical and hydrometallurgical processes. It does not require thorough dissolution of metal elements in the cathode materials. In response to the failure problems such as crystal distortion, loss of active lithium, and surface impurities in retired cathode materials, it repairs performance through processes such as lithium supplementation, crystal structure reconstruction, and surface modification. It has the advantages of short process, low energy consumption, and small carbon footprint, reducing carbon emissions by about 70% compared to traditional hydrometallurgical processes. It is a cutting-edge research direction for green recycling of waste lithium batteries [8]. Its core goal is to enable the performance of regenerated cathode materials to approach that of pristine materials, reduce recycling costs, and promote the closed-loop development of lithium batteries throughout their entire lifecycle.

Solid phase sintering regeneration method is currently the most mature direct regeneration technology, which involves mixing waste lithium iron phosphate cathode materials with lithium salts such as lithium carbonate in proportion, and reconstructing the olivine crystal structure through high-temperature treatment [9]. With optimized technical parameters, the cycle life of regenerated lithium iron phosphate can reach over 2000 cycles, with a discharge capacity difference of nearly 5% compared to the original material, and its performance is basically equivalent to that of commercial lithium iron phosphate electrodes. In recent years, breakthrough progress has been made in ultra-fast regeneration technology: a research team from Tsinghua University published in Advanced Materials in 2025 achieved single-step ultra-fast upgrade regeneration of waste lithium iron phosphate, using green reagents to

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synergistically repair cathode defects. The initial discharge specific capacity of the regenerated material at 0.1 C rate reaches 163 mAh g^{-1}, and its capacity retention rate remains at 93.5% after 500 cycles at 1 C rate; The team will also upgrade waste lithium iron phosphate and manganese-rich cathode materials to lithium manganese iron phosphate. The energy density of the regenerated product will increase by 6.7% compared to traditional LFP, and the economic benefits will increase by 2–4 times compared to traditional regeneration technology. The flash electric heating regeneration technology developed by Rice University can quickly reconstruct the crystal structure of lithium cobalt oxide. The metal recovery rate of the regenerated material reaches 98%, and the electrochemical performance is close to that of commercial pristine products, with some indicators even better. The Joule heating high-temperature shock strategy proposed by the Guangzhou Institute of Energy, Chinese Academy of Sciences, can activate and repair cathode materials within 1 second, with a lithium leaching rate of over 99%, significantly improving regeneration efficiency, and achieving synergistic high-value recovery of retired battery cathode materials and leaching residues.

At present, the cost of direct regeneration technology is about 60%–70% of that of pristine cathode materials, and it can be adapted to various cathode material systems such as lithium iron phosphate and lithium manganese oxide. It is in the transition stage from pilot-scale testing to industrial application. With the promotion of process standardization, this technology will become the mainstream development direction for high-value regeneration of cathode materials.

6. Challenges and Trends in the Development of Green Recycling Technology

Direct regeneration technology is different from the "disassembly and extraction" mode of pyrometallurgical and hydrometallurgical processes. It does not require complete dissociation of the cathode metal components and can achieve performance repair through lithium supplementation, crystal reconstruction, and surface modification to address degradation mechanisms such as retired cathode crystal distortion, active lithium loss, and surface impurities. It has the advantages of simple process, low energy consumption, and low carbon emissions, reducing carbon emissions by about 70% compared to traditional hydrometallurgical processes. It is an important direction for green recycling of waste lithium batteries [10]. This technology aims to make the performance of regenerated cathode materials approach that of native materials, reduce recycling costs, and promote closed-loop cycling of lithium batteries throughout their entire lifecycle.

Solid phase sintering regeneration is the most mature direct regeneration process, which involves mixing waste lithium iron phosphate with lithium salts and subjecting it to high-temperature heat treatment to reconstruct the olivine crystal structure. Under optimized processes, the cycle life of regenerated lithium iron phosphate exceeds 2000 cycles, with a discharge capacity difference of nearly 5% compared to the original material, which is comparable to the performance of commercial electrodes. The development of ultrafast regeneration technology has been significant in recent years: the Zhou Guangmin Team from Tsinghua University and the team of academician Cheng Huiming from the Chinese Academy of Sciences published their results in Nature Protocols in 2025, using Joule heating non-equilibrium repair to achieve one-step upgrading of waste lithium manganese oxide to high-pressure nickel lithium manganese oxide, with better performance than commercial materials; Related research can also upgrade lithium iron phosphate with manganese-rich materials, increasing energy density by 6.7% compared to traditional LFP and increasing economic benefits by 2–4 times. Rice University's flash electric heating technology can quickly reconstruct lithium cobalt oxide crystals, with a metal recovery rate of 98%; The Joule thermal

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shock strategy of Guangzhou Institute of Energy, Chinese Academy of Sciences can complete cathode activation and repair within 1 second, with a lithium leaching rate exceeding 99%.

The cost of direct regeneration technology is approximately 60%–70% of that of pristine cathode materials, and it can be adapted to various cathode material systems, shifting from pilot testing to industrialization. CATL (Contemporary Amperex Technology Co., Limited)'s direct regeneration technology achieves a nickel cobalt manganese recovery rate of 99.6% and a lithium recovery rate of 96.5%, with carbon emissions reduced by more than 60% compared to wet processes. The EU's New Battery Law will implement battery passports in February 2027, and China will simultaneously build a digital traceability system for power batteries. Relevant national and group standards are being continuously improved. According to industry associations' predictions, the market size of direct regeneration technology will reach 30 billion RMB by 2030, becoming the mainstream technology for high-value regeneration of cathode materials.

7. Conclusion

In summary, the green recycling and regeneration technology for the cathode materials of waste lithium batteries is the core approach to alleviating the scarcity of strategic mineral resources, reducing environmental pollution, and promoting the sustainable development of the new energy industry. Research has shown that the three mainstream recycling technologies of pyrometallurgy, hydrometallurgy, and direct regeneration have all achieved green upgrades: pyrometallurgy reduces energy consumption and exhaust emissions through improvements such as low-temperature roasting and exhaust gas resource utilization, while hydrometallurgy relies on green leaching agent substitution and process optimization to achieve highly selective recovery of valuable metals. Direct regeneration achieves high-value repair of cathode materials through short processes and low-carbon advantages. These three technologies form a complementary technical system, addressing the drawbacks of high pollution, low efficiency, and high energy consumption in traditional recycling processes, and providing support for the industry's green transformation.

There are still obvious limitations in the current industry: a low level of technological integration, as well as the difficulty in efficient collaborative treatment of cathode materials from different systems; The incomplete industrial support system and inconsistent performance evaluation standards for regenerated materials have limited the large-scale promotion of green recycling technology and hindered its industrial value release.

In the future, it is necessary to focus on the core development directions and strengthen collaborative innovation between industry, academia and research, overcome key technologies such as multi-system cathode material adaptation, green leaching reagent research and development, and intelligent recycling equipment upgrading, and facilitate the industrialization of research achievements; A sound recycling network for waste lithium batteries should be established, and the entire recycling process should be standardized, and ensure the supply of raw materials; Improve the performance standards of regenerated cathode materials and enhance the market competitiveness of products. With the tightening of environmental policies and the maturity of technological iteration, this technology will develop towards low-carbonization, high-value, and intelligentization. The cost-effectiveness of regenerated materials will continue to improve, and it is expected to replace primary minerals, realizing the large-scale, green, and high-value recycling of waste lithium battery cathode materials, laying a solid resource foundation for the sustainable development of the new energy industry, and thus supporting China in achieving its Dual Carbon Goals.

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