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Review

Advances in Recycling Technologies of Critical Metals and Resources from Cathodes and Anodes in Spent Lithium-Ion Batteries

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Abstract: With the rapid economic development and the continuous growth in the demand for new energy vehicles and energy storage systems, a significant number of waste lithium-ion batteries are expected to enter the market in the future. Effectively managing the processing and recycling of these batteries to minimize environmental pollution is a major challenge currently facing the lithium-ion battery industry. This paper analyzes and compares the recycling strategies for different components of lithium-ion batteries, providing a summary of the main types of batteries, existing technologies at various pretreatment stages, and recycling techniques for valuable resources such as heavy metals and graphite. Currently, pyrometallurgy and hydrometallurgy processes have matured; however, their high energy consumption and pollution levels conflict with the principles of the current green economy. As a result, innovative technologies have emerged, aiming to reduce energy consumption while achieving high recovery rates and minimizing the environmental impact. Nevertheless, most of these technologies are currently limited to the laboratory scale and are not yet suitable for large-scale application.

Keywords: lithium-ion batteries; technologies; recovery; resources

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1. Introduction

One of the main contradictions facing contemporary society is the rapid growth in energy demand coupled with the diminishing availability of traditional non-renewable energy sources. Amid China's carbon reduction initiatives, electric vehicles (EVs) are gradually being promoted as a widely implemented mode of transportation for reducing greenhouse gas emissions and lessen the dependence on fossil fuels [1]. Lithium-ion batteries (LIBs), due to their superior electrochemical performance and environmental friendliness, are widely used not only in electric vehicles and other everyday electronic devices (Figure 1a) but also in satellites, energy storage systems, and aerospace. As demand and production rise each year, the pursuit of carbon neutrality is expected to trigger exponential growth in the demand for lithium batteries. According to recent data, in 2023, China's production of new energy vehicles reached 9.587 million units, accounting for 64.8% of global sales. The production of lithium-ion batteries reached 778.1 GWh, a year-on-year increase of 42.5% [2], with global demand for lithium-ion batteries expected to continue rising further in the coming decade. With the rapid development of the electric

vehicle market, we may face significant challenges in handling waste lithium-ion batteries in the coming years. The substantial increase in retired electric vehicle batteries will have a profound impact on resource conservation and environmental protection, necessitating immediate and effective measures. Thus, focusing on renewable energy sources is essential for achieving current and future carbon reduction goals.

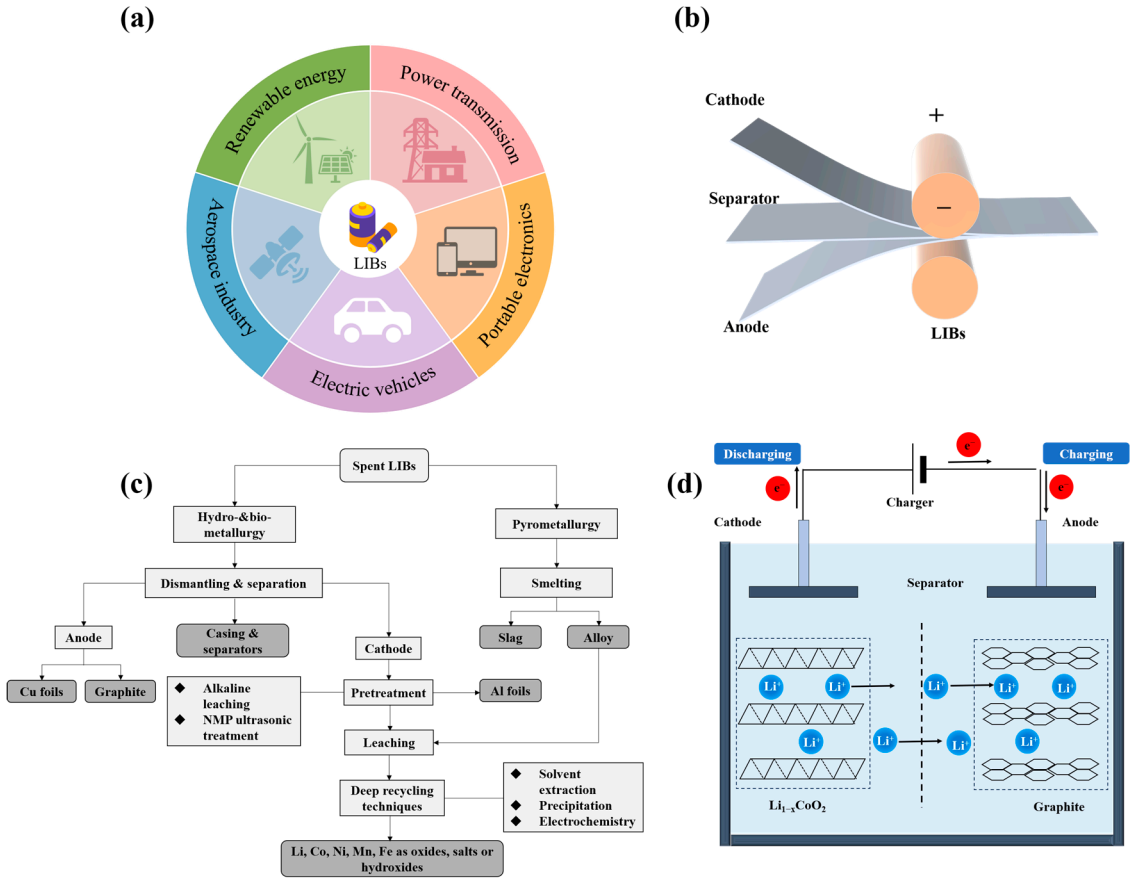


Figure 1. (**a**) Applications of lithium-ion batteries; (**b**) The shape and components of some Li-ion battery configurations; (**c**) Flow-chart showing the typical recycling process; (**d**) Schematic diagram of the LIB working principle.

Generally, the lifespan of lithium-ion batteries is around 5–8 years. With increasing demand, a significant number of these batteries are expected to be retired in the coming years [3]. As major participants in the global electric vehicle market, Europe, China, and the United States have each formulated policies to manage retired batteries, protect the environment, and recycle renewable resources [4,5]. As illustrated in Figure 1b, the main components of lithium-ion batteries include the cathode, anode, metal shell, and separator. These batteries contain several high-value metals (e.g., Li, Ni, Mn, Co, Cu, Al, etc.) [6,7]. Improper recycling can pollute soil and groundwater during landfilling and lead to economic losses, further aggravating resource scarcity issues [8,9]. Previous studies indicate that researchers have successfully recovered valuable metals from end-of-life LIBs using pyrometallurgical and hydrometallurgical processes, as raw materials for new batteries or as functional materials for various applications [10]. As shown in Figure 1c, the recycling flowchart illustrates several common recycling methods. Despite variations among manufacturers, cathode materials, applications, and shapes of lithium-ion batteries, their working mechanisms remain fundamentally similar. The electrochemical working mechanism of lithium-ion batteries is shown in Figure 1d. For instance, the working mechanism of $LiCoO_2$ is as follows:

During the charging process, lithiumions are extracted from $LiCoO_2$, and Co^{3+} is oxidized to Co^{4+}. At the anode, lithium ions and electrons released from the cathode react to produce lithium carbide (LiC_6). During discharge, lithium ions deintercalate from the graphite anode and intercalate into the $LiCoO_2$ cathode, resulting in the reduction of Co^{4+} to Co^{3+}. The chemical reactions of the charge–discharge process are shown in Table 1:

Table 1. The charge–discharge process of $Li_{1-x}CoO_2$.

Charge–Discharge Process		
Charging reaction	Cathode	$LiCoO_2 = Li_{1-x}CoO_2 + xLi^+ + xe^-$
	Anode	$6C + xLi^+ + xe^- = Li_xC_6$
Discharging reaction	Cathode	$Li_{1-x}CoO_2 + xLi^+ + xe^- = LiCoO_2$
	Anode	$Li_xC_6 = 6C + xLi^+ + xe^-$

Although many reviews have explored the recovery of metal resources from waste batteries, most of them focus on the partial recovery of SLIBs, lacking a systematic summary of recovery methods for metals and other resources in waste batteries. This paper not only reviews the progress of research on recovering metals and other resources from various types of waste batteries but also systematically summarizes related technical methods, including the recovery of metal resources from cathodes (e.g., lithium, cobalt, nickel, manganese) and graphite from anodes, aiming to improve the overall recovery efficiency of lithium-ion batteries. Finally, from an application perspective, the paper summarizes the development of various recovery processes, the performance of recovered products, and the environmental impact of the recovery process. Additionally, it highlights the challenges currently faced in the field of recovery and predicts future development trends.

2. Composition and Pretreatment of LIBs

2.1. Composition

Lithium-ion batteries consist of five main components: the cathode, anode, separator, electrolyte, and metal shell. Table 2 shows the proportional composition and associated hazards of each component.

Table 2. Component of LIBs [11–13].

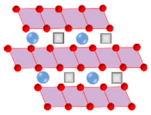
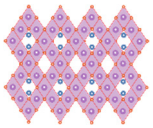
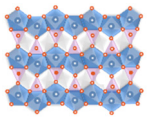
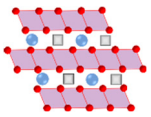
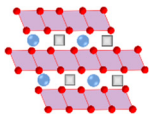
Cell Components	Chemical Composition		wt., %	Hazards
Cathode	Metal oxide	85% metal oxide powders ($LiNiO_2$, $LiMn_2O_4$, $LiCoO_2$, etc.) and 10% binder		Reacts with acidic and ammoniacal organic
	Aluminum	Al(Current collector foil)	5–8	
	Binder	Usually PVDF	1–2	
Anode	Copper	Cu (Current collector foil)	8–10	Reaction with oxidizers produces carbon oxides, contributing to the greenhouse effect
	Binder	Usually PVDF	1–2	
	Graphite	-	15–17	

Table 2. *Cont.*

Cell Components	Chemical Composition		wt., %	Hazards
Electrolyte	Li salts	$LiPF_6$, $LiAsF_6$, $LiClO_4$, $LiBF_4$	10–15	Both $LiBF_4$ and $LiBF_6$ are highly corrosive, decomposing to release fluoride pollutants. Their corrosive and volatile nature poses risks to the respiratory system and other parts of the body.
	Organic solvents	DMC-EC, PC-DME, BL-THF		
Casing		Fe–Ni alloy	20–26	-
		Al	10	
Other		Positive and negative electrode leads, center pin, insulating materials, safety valve, PTC (Positive Temperature Coefficient terminal)	18–20	-

The cathode is composed of carbon black, polymer binder, and lithium transition metal oxide coated on aluminum foil. Based on their crystal structure, lithium-ion batteries can be classified into layered structures ($LiCoO_2$, $LiNiMnCoO_2$, $LiNiCoAlO_2$), spinel structures ($LiMn_2O_4$), and olivine structures ($LiFePO_4$). The layered structure is characterized by a two-dimensional planar form, offering high ionic mobility and energy density. Spinel structures feature three-dimensional lithium-ion migration channels, enabling faster ion diffusion rates, but have relatively shorter cycle lives. Olivine structures, on the other hand, have one-dimensional ion diffusion pathways and are structurally stable but exhibit low conductivity, typically improved through carbon coating or nanostructuring [14,15]. A comprehensive comparison of commonly used lithium-ion battery cathode materials is shown in Table 3. Tesla Roadster originally utilized LCO batteries; however, shortly thereafter, the majority of EV manufacturers opted for NCM or NCA batteries due to their extended cycle lifespan. The application ratio of NCM materials in power lithium-ion batteries has shown consistent growth in recent years. By 2020, NCM lithium-ion batteries were anticipated to produce 71.6 GWh, representing more than 70% of the power battery sector, with predictions of them dominating the global market share by 2025 [16,17]. While the dependence of electrode materials on strategic resources like cobalt has diminished, the surging market demand continues to necessitate increased resource availability. The anode consists of graphite coated on copper foil and a polymer binder [18]. Common binders include polyvinylidene fluoride (PVDF) and polytetrafluoroethylene (PTFE). The binder system also includes solvents such as N-methyl-2-pyrrolidone (NMP) or N,N-dimethylformamide (DMF), which help mix the binder with conductive and active materials. Commercial separators are typically made of polypropylene (PP) and polyethylene (PE) [19]. Their function is to isolate the cathode from the anode, preventing the free flow of electrons in the battery and thus avoiding short circuits. Meanwhile, different lithium-ion batteries use various electrolytes. For instance, lithium-ion batteries commonly employ lithium salts such as lithium hexafluorophosphate ($LiPF_6$), lithium tetrafluoroborate ($LiBF_4$), lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), etc. These electrolytes dissolve in nonpolar propylene carbonate solvents, such as ethylene carbonate/dimethyl carbonate (EC/DMC) [20].

Table 3. Comprehensive comparisons of cathode materials [2,16].

Cathode Types	LCO	LMO	LFP	NCA	NCM
Chemical formula	$LiCoO_2$	$LiMn_2O_4$	$LiFePO_4$	$LiNi_xCo_yAl_zO_2$	$LiNi_xCo_yMn_zO_2$
Structure					
Market share	Dumped	Small	Growing	Steady	Main force
Comments	Low safety, high cost, medium performance	Medium safety, low cost, medium energy density, low lifetime	Good safety, low cost, high thermal stability, medium energy density	Medium safety, medium cost, high energy density	Medium safety, medium cost, high energy density, high lifetime

2.2. Pretreatment

2.2.1. Discharge Process

Some discarded lithium-ion batteries still carry a certain residual charge. The disassembly or crushing of lithium-ion batteries can result in direct contact between the anode and cathode materials, increasing the risk of short circuits, fires, or even explosions. Therefore, discharging discarded lithium-ion batteries is a crucial step to ensure subsequent graphite separation. Several discharge techniques have been created, such as solution discharge, low-temperature cryogenic discharge, solid-state conductor discharge, and thermal deactivation. The comparison of different discharging technologies is presented in Table 4. Xiao et al. proposed an O_2 control mechanism to explain the battery discharge process in a $MnSO_4$ solution, where manganese (II) can be in situ transformed to form an isolation layer, reducing oxygen-anode contact and effectively preventing galvanic corrosion and organic leakage. In summary, this method achieves a more environmentally friendly lithium-ion battery discharge approach [21]. However, the waste liquid generated from the discharge using salt solutions still requires further treatment. Zheng et al. identified the key characteristic of a physical discharge medium, namely, graphite powder. The discharge rate in graphite powder is relatively mild [22]. Cryogenic freezing has been used in several studies, but its efficiency is rarely explored due to high equipment demands, high initial construction costs, and limited applicability for large-scale use. Moreover, as the number of waste lithium-ion batteries continues to rise, there is still significant research potential regarding the environmental impacts of different discharge methods.

Table 4. The comparison of different discharging processes.

Approach	Discharge Medium	Advantage	Disadvantage	Ref.
Salt solution discharge	Salt solution (including NaCl, KCl, $FeSO_4$, $MnSO_4$ and $ZnSO_4$ solution, etc.)	Cheap, easy to obtain, controllable	Low discharge speed; metals and electrolytes leak into the solution; gaseous pollutants	[21]
Cryogenic freezing	Liquid nitrogen	Environmentally friendly	High capital and equipment requirements	[23]
Solid electrical conductors	Cu powder or graphite	Fast discharge speed, no aqueous pollution	Unsafe, potential dust pollution, unstable	[24]
Thermal deactivation	None	Elimination of the potential gaseous pollutants	High-temperature requirements	[25]

2.2.2. Disassembly and Separation

Dismantling and separation are generally categorized into manual and mechanical methods. Laboratories typically use manual dismantling methods. Although manual disassembly achieves high efficiency and purity in separation, the labor-intensive process and notable safety concerns—especially in handling batteries with hazardous materials like LiPF6, HF, and leftover lithium—render it inappropriate for large-scale industrial applications [13,26]. In comparison, mechanical dismantling provides an effective, safe, and scalable alternative that is appropriate for waste electrodes or entire batteries [27]. On one hand, mechanical dismantling is cost-effective, easy to operate, highly efficient, and capable of processing large volumes, enabling the effective release of battery electrode materials and valuable components; on the other hand, the particle size distribution and the degree of release in the crushed products significantly impact the following recovery efforts. The process of mechanical dismantling consists of various steps, such as crushing, screening, flotation, and magnetic separation. Currently, the crushing techniques used in production are divided into dry and wet methods. Dry crushing directly processes discarded lithium-ion batteries in a water-free environment, separating components. However, the crushing process can result in the release of gases [28]. It has been reported that carbon dioxide, ethyl methyl carbonate (EMC), and dimethyl carbonate (DMC) are the main active gases produced during the crushing process. Wet crushing, on the other hand, is performed in a solution, converting the batteries into a non-reactive state and decreasing their reactivity with electrolytes [29]. Considering the goal of harmless treatment, wet crushing has been shown to be more effective in reducing the toxic emissions.

The further processing and separation of dismantled and crushed raw materials can produce higher-grade materials. Various separation processes are frequently employed, including gravity separation, electrostatic separation, eddy current separation, and froth flotation, to remove external polymers and casings, concentrating the internal metallic components [30]. After crushing, the particle sizes of components such as electrode coatings, graphite, and metal oxides vary, allowing for separation using the gravity screening method, which is easy to operate and low-cost, though its separation efficiency is limited. Due to the interpenetration of different LIB components, such as the close bonding between PVDF binders and current collectors, achieving complete separation through simple screening remains challenging. In addition to metals, the used LIBs also contain non-metallic components, such as polymers. Electrostatic separation is an effective method for separating metals from non-metals [31]. Eddy current separation is a method that pre-sorts different materials based on changes in physical properties; Elements with specific conductivities can be separated using varying sizes of eddy currents, significantly enhancing the recovery efficiency of waste lithium-ion batteries [32]. Flotation is a physical process that selectively separates hydrophobic materials from hydrophilic ones by exploiting differences in material wettability and the effects of collectors, frothers, and modifiers. Flotation can effectively enhance the purity of recovered metals, reducing impurities and thereby decreasing the refining requirements for subsequent smelting. This method has been widely applied in the recovery of cobalt and nickel. Compared to other chemical separation methods, flotation is a relatively eco-friendly separation technology that primarily relies on physical properties, generates minimal chemical waste, and imposes a lower burden on waste liquid treatment, thus having a reduced environmental impact [33].

3. Recycling of Anode Electrode Materials

In recent years, anode recycling has gradually gained attention, although it has yet to reach the prevalence of cathode recycling. Currently, the anode is becoming increasingly important in lithium-ion battery recycling and can be made from various materials, such as natural or synthetic graphite, carbon-based materials, or even silicon. Among these materials, graphite is the most widely used option in commercial applications due to its excellent conductivity, stability, and low cost [34]. Recovering graphite from anode waste serves as an efficient means of acquiring high-quality graphite and plays a vital role in reducing the costs associated with producing battery-grade graphite. This approach is essential for preventing environmental pollution, lowering production costs, and advancing sustainable development. Therefore, it is imperative to separate and recover graphite from used lithium-ion batteries.

However, a key challenge in anode recycling is that graphite particles are easily contaminated by metals and other impurities during the recycling process, which can impair material performance. Therefore, exploring cost-effective recycling methods is crucial for the regeneration of anodes in spent lithium-ion batteries. Currently, the primary anode recycling methods include mechanical–physical processes, acid leaching, pyrolysis, and some novel methods, such as electrolysis, subcritical CO_2 extraction, and plasma aqueous solution methods. Table 5 compares the main graphite recycling methods and their electrochemical properties. Yang et al. first employed a roasting technique to eliminate organic materials (such as binders and SEI films) from waste graphite, followed by acid leaching to further extract impurities [35]. With an initial discharge capacity of 591.0 mAh/g, the regenerated graphite maintained a cycling stability of 97.9% over the course of 100 cycles. However, this method may lead to the structural degradation of carbon materials, affecting the performance while releasing harmful gases or by-products that can cause environmental issues. In their study, Ma et al. applied a treatment to waste graphite using a mixture of 5 mol/L H_2SO_4 and 35.0 *w*/*w*% H_2O_2 and subsequently roasted it with NaOH at 500 °C [36]. The results showed that impurities were substantially removed, with the theoretical capacity of regenerated graphite reaching 377.7 mAh/g, though the graphite yield remained at around 60%, possibly requiring further optimization. Traditional regeneration techniques face obstacles such as a reliance on hazardous chemicals, significant energy use, and complications with large-scale applications. Fan et al. improved the electrochemical performance of regenerated graphite through microwave-assisted calcination, adjusting parameters such as duration and power to achieve optimal outcomes [37]. Cao et al. introduced an electrolysis technique to separate anode materials and copper foil from LIBs [38]. Through the optimization of various reaction conditions, the effective separation of copper foil and anode materials was achieved, with the purity of the regenerated graphite reaching 95%. In conclusion, compared to conventional methods of graphite production and recycling, innovative techniques like electrolysis and microwave assistance substantially lower energy consumption and greenhouse gas emissions, making graphite recycling more eco-friendly and economical [39]. Characterized by being chemical-free, efficient, and scalable, these methods represent significant progress in the area of graphite recycling.

Table 5. Comparison of different regeneration methods of waste graphite and their electrochemical properties.

Sample	Method		Performance	Ref.
Waste graphite	Chemical	Citric acid leaching	Initial charge capacity and retention rate are 468.3 mA h/g at 0.5 C and 68.3%	[40]
	Physico-chemical	H_2SO_4 curing anaerobic calcination	Initial charge capacity and retention rate are 349 mA h/g at 0.1 C and 98.8%	[41]
		H_2SO_4 leaching– calcination at 3000 °C—pitch coating	Specific capacity of 344 mA h/g at 0.2 C	[42]
		Sulfuric acid curing–leaching combined with high-temperature calcination	Initial charge capacity and retention rate are 349 mA h/g and 99.4% after 100 cycles at 0.1 C	[41]
		H_2SO_4 leaching–heat- treated at 900 °C for 2 h	Initial specific capacity (358.1 mA h/g at 0.1 C) and cycle stability of 98.8% after 100 cycles	[43]
		5 mol/L H_2SO_4, 35.0 *w*/*w*% H_2O_2, and NaOH at 500 °C	Initial charge capacity and retention rate are 431.9 mA h/g and 84.6% after 100 cycles at 0.2 C	[36]
		Sulfuric acid curing–leaching and microwave-assisted calcination	Initial charge capacity and retention rate are 424.8 mA h/g and 98.3% after 60 cycles at 0.1 C	[37]
		Two-stage calcination and HCl solution	Initial charge capacity and retention rate are 591 mA h/g and 97.9% after 100 cycles at 1.0 C	[35]
	Physical	Electrochemical method	First discharge and charge specific capacities at a rate of 0.1 C are 427.81 mA h/g and 350.47 mA h/g	[38]
		Microwave-irradiation time of 15 s	Reversible capacity of ≥400 mA h/g at 0.1 mA h/g and an initial coulombic efficiency of 84%	[44]
		Roasting at 1600 °C for 1 h	Reversible capacity of 235 mA h/g at 1 C after 500 cycles	[45]

4. Recycling of Cathode Electrode Materials

The cathode of lithium-ion batteries contains various metal components, necessitating multistep processes to selectively separate these metals. Pyrometallurgy and hydrometallurgy are traditionally used for cathode metal leaching and can achieve efficient separation through synergistic effects. To achieve energy conservation and emission reduction goals, direct recycling methods for lithium-ion batteries have advanced in recent years [46]. With continuous technological advancements, several novel methods for metal separation have emerged [47]. Figure 2 presents a schematic diagram of pyrometallurgy, hydrometallurgy, and direct recycling processes.

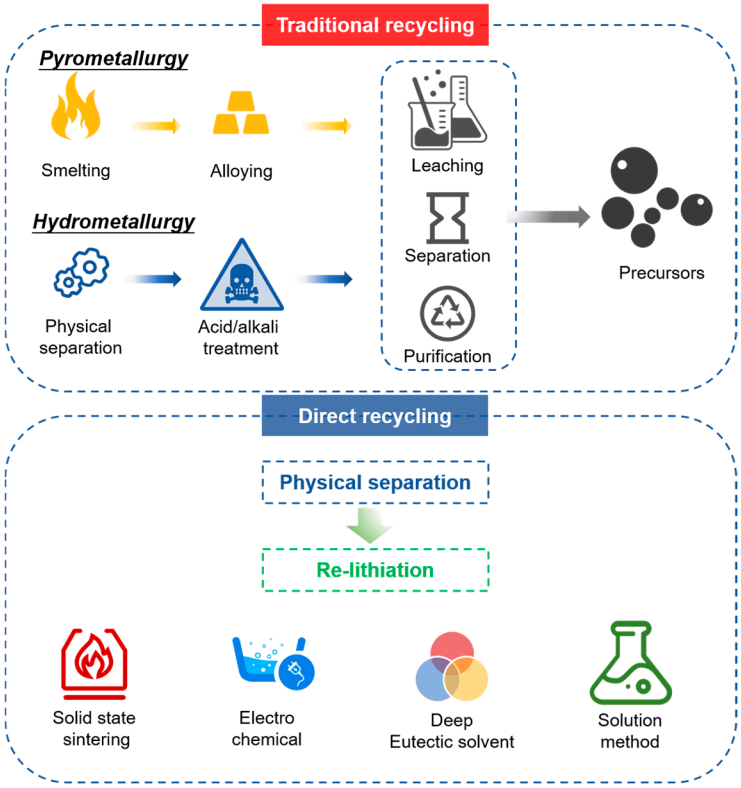


Figure 2. Schematic diagrams of pyrometallurgy, hydrometallurgy, and direct recovery processes [48].

4.1. Hydrometallurgy

Hydrometallurgy is a metallurgical process that extracts metals from ores, waste, or battery materials through liquid-phase reactions. It is widely used in lithium-ion battery recycling, especially for extracting valuable metals such as lithium, cobalt, and nickel from electrode materials [49]. High-value elements are separated and recovered from the leachate using techniques such as solvent extraction, electrolysis, precipitation, and ion exchange. Compared to pyrometallurgy, hydrometallurgy offers advantages such as lower emissions, reduced energy consumption, greater flexibility, and higher purity and recovery rates [50,51]. Consequently, hydrometallurgy is regarded as more suitable for recycling end-of-life LIBs. However, this approach also presents challenges, such as stringent recovery requirements, considerable upfront investment, and the generation of significant amounts of acidic and heavy-metal-rich wastewater, all of which contribute to increased costs and complexity in the recycling process [52].

4.1.1. Leaching

Acids or bases are used to convert metal oxides or metals in battery materials into soluble compounds for dissolution in the liquid phase. The subsequent leachate must undergo further processing to separate metal ions and produce the final products. Acidic, alkaline, and bio-leaching methods are typically used to extract metals from discarded lithium-ion batteries (LIBs). The details are shown in Table 6.

Table 6. Chemical leaching of spent LiBs.

Type	Reagents	Target Metals	Conditions	Efficiency	Ref.
Inorganicacid	H_2SO_4 with H_2O_2 and glutaric acid	Co, Li, Ni	50–90 °C, 500 rpm for 120 min	87.85% Co, 99.91% Li, and 91.46% Ni were recovered	[53]
	H_2SO_4 and sucrose	Co, Li	95 °C, 120 min, 25 g/L	100% Li and Co 96%	[54]
	HCl and H_2O_2	Co, Li	75 °C, 30 min, 5 g/L	93% Li and Co	[55]
	Na_2SO_3	Co, Ni, Mn	120 °C, 480 min, 2 g/L	93.1% Ni, 92.84% Co, and 90.2% Mn	[56]
	$H_4P_2O_7$	Fe, Li	25 °C, 300 min, 100 g/L	100% Fe and 97.98% Li	[57]
Organic acid	Citric acid and H_2O_2	Co, Li, Ni, Al, Mn	95 °C, 20 min, 20 g/L	93% Al, 90% Co, 96% Li, 94% Mn, and 94% Ni	[58]
	Lemon juice and H_2O_2	Li, Cu, Al	20 °C, 90 min, 67 g/L	94.8% Li, 96.9% Cu, and 47.2% Al	[59]
	Citric acid and H_2O_2	Co, Li, Ni, Mn	95 °C, 120 min	96% Li, 87% Co, 93.5% Ni, and 90.5% Mn	[60]
	DL-malic acid and H_2O_2	Co, Li, Ni, Mn	90 °C, 30 min, 40 g/L	98.9% Li, 94.3% Co, 95.1% Ni, and 96.4% Mn	[61]
	CH_3SO_3H	Li, Fe	90 min, 80 g/L	94% Li and 95% Fe	[62]
Alkali Leaching	$(NH_4)_2SO_3$ NH_3 and NH_4HCO_3	Li, Co, Ni	60 °C, 180 min, 20 g/L	60.53% Li, 80.99% Co, and 96.32% Ni	[63]
	$(NH_4)_2SO_4$, NH_3 and Na_2SO_4	Li, Co	80 °C, 300 min, 40 g/L	96% Li and 81% Co	[64]
	NaOH	Li, Al, Co	85 °C,150 min, 30 g/L	80.76% Li, 100% Al, and 93% Co	[65]
Bio-leaching	Acidithiobacillus ferrooxidans	Ni, Mn, Co, Li	72 h, 100 g/L	90% Ni; 92% Mn; 82% Co; 89% Li.	[66]
	H_2SO_4 and antibiotic bacteria residues (mainly CaC_2O_4 hydrates)	Ni, Mn Co, Li	30 mL/g, 363 K for 2.5 h	99.9% Li, 98.5% Co, 99.0% Mn, and 99.6% Ni	[67]
	Acidithiobacillus ferrooxidans	Ni, Mn, Co, Li	100 g/L, 30 °C 360 min, pH = 6	89.9% Li, 90.4% Co, 91.8% Mn, and 85.5% Ni	[68]

Acid leaching: Acid leaching encompasses two methods: inorganic acid leaching and organic acid leaching. Common inorganic acids include HCl [69,70], H_2SO_4 [71,72], HNO_3 [73], and H_3PO_4 [74]. Inorganic acids can complete leaching in a short time and are suitable for processing various electrode materials (such as complex compounds like $LiCoO_2$ and $LiNiMnCoO_2$), offering high leaching rates and potential for large-scale industrial applications. However, this leaching technique also presents challenges, such as corrosion, waste acid contamination, and the emission of toxic gases (like NO_x and Cl_2), which increase the environmental burden. Barik and He et al. achieved over 99% leaching

efficiency by utilizing HCl and H_2SO_4-H_2O_2 systems for dissolving metals such as Co, Mn, and Li (Barik et al., 2017, He et al., 2017). Organic acid leaching is a developing technology for processing waste lithium-ion batteries. Common organic acids include citric acid [75], ascorbic acid [76], oxalic acid [77], lactic acid [78], acetic acid (Li et al., 2018), and formic acid [79]. Compared to inorganic acids, organic acid leaching offers advantages such as being more environmentally friendly, easily degradable, and producing less secondary emissions; however, its high costs and slower leaching rates limit its industrial application [80,81]. Sun et al. employed oxalate as both a leaching and precipitation agent to extract metals from lithium-ion batteries, achieving extraction rates exceeding 98% for both Li and Co, with an extraction efficiency of 90% [82]. Gao et al. incorporated formic acid into their process, selectively leaching lithium ions into the solution while precipitating other metals for precursor production. After removing metal residues, Li_2CO_3 with a purity of 99.9% was produced [79].

Alkali Leaching: Alkaline leaching is a process that uses strong bases (like NaOH and Na_2CO_3) to dissolve or convert certain metal oxides in battery materials into soluble complexes for the extraction of specific metals from discarded batteries. Zheng et al. used ammonia and ammonium sulfate as leaching agents, along with sulfite as a reducing agent, attaining overall leaching efficiencies of 94.8% for nickel, 88.4% for cobalt, and 96.7% for lithium [64]. Alkaline (ammonia) leaching offers higher recovery rates, shorter processing times, and the continuous extraction of valuable metals, making it suitable for the large-scale processing of cathode waste. Alkaline leaching primarily targets metals that are soluble in alkali, such as aluminum (Al) and lithium (Li), while the solubility of other metals, such as cobalt and nickel, is relatively low, which limits the recovery targets. Therefore, alkaline leaching is often combined with acid leaching processes [12,83].

Bioleaching: Bioleaching is a technology that utilizes acidic substances produced by microbial metabolism or redox reactions to convert metals in waste batteries into soluble ions for extraction. Do et al. conducted the bioleaching of waste NMC using ferrous oxidizing microbes, recovering 90.4% of Co, 89.9% of Li, 85.5% of Ni, and 91.8% of Mn [68]. Bioleaching shows great promise in the field of battery recycling due to its environmental friendliness, low energy consumption, and strong adaptability. Typically, bacteria produce inorganic acids, while fungi generate organic acids. However, due to the slow reaction rate, it typically takes several days to weeks to achieve optimal leaching results, which limits its large-scale industrial application [84,85].

4.1.2. Separation and Recovery of Valuable Elements from Leaching Solutions

The leachate after hydrometallurgy typically contains various metal ions and trace impurities, necessitating separation and purification to achieve the recycling of metal resources. Previously introduced methods include solvent extraction [86,87], ion exchange (Haruna et al., 2024), chemical precipitation [88], electrochemical deposition (Pei et al., 2024), and sol-gel processes, which are employed for separating metal ions from leachate or removing impurities. The combination of solvent extraction and chemical precipitation is regarded as an efficient method to separate and recover metals from leach solutions [80]. Specific research is presented in Table 7. The effect of pH on the metal separation process in hydrometallurgical leachate is highly significant. It not only determines the speciation of metal ions but also affects the efficiency of key processes such as precipitation, extraction, and adsorption [89]. Therefore, optimizing the pH value is a crucial step in designing separation processes to improve the recovery rate and selectivity of target metals [90]. However, this aspect has received relatively little attention so far, making it one of the key issues that need to be prioritized in future studies on recycling waste lithium-ion batteries.

Solvent Extraction: Solvent extraction involves selecting a mixture of one or more organic solvents as the extracting agent, forming coordination complexes with desired metal ions, and then transferring these metal ion complexes to another organic solvent for the separation and purification of different metals. The special selectivity and high separation efficiency of the extracting agents for different metal ions are key reasons for their widespread application in hydrometallurgy. Solvent extraction offers advantages such as high product purity and short reaction times; however, its complex operation and high solvent costs limit its application. Commonly used extracting agents include di(2-ethylhexyl) phosphoric acid (D2EHPA) [91], Cyanex 272 [92], and 2-ethylhexyl phosphoric acid mono-2-ethylhexyl ester (PC-88A) [93]. Different types of extracting agents have unique characteristics, and suitable combinations of extracting agents are typically selected based on the types of metals and processing conditions in practical applications. Additionally, the synergistic effects among extracting agents can also be utilized to enhance extraction efficiency [94,95]. Mahmoudi et al. [96] compared the extraction and separation efficiencies of Li^+ and Co^{2+} using Cyanex 272, D2EHPA, and their mixtures. The results indicate that Cyanex 272 and D2EHPA exhibit significant synergistic extraction effects, with an optimal ratio of 5:1.

Chemical Precipitation: This method of chemical precipitation is used to separate dissolved metal ions into precipitates for metal recovery. Chemical precipitation operates on the principle of different solubilities of metal compounds under certain temperature and pH conditions, which requires modifications during the precipitation process. Commonly used precipitating agents include sodium hydroxide (NaOH) (Liu et al., 2019b), sodium carbonate (Na_2CO_3) [97], sodium phosphate (Na_3PO_4) [98], oxalic acid ($H_2C_2O_4$) [99], and oxalates [76]. Due to its efficiency, cost-effectiveness, and environmental friendliness, the chemical precipitation method has been widely applied in the area of metal recovery, particularly in lithium-ion batteries.

Deep eutectic solvents: DES are green solvents characterized by high renewability, biodegradability, and recyclability [100]. Additionally, DES possess low toxicity, low flammability, low solubility, and low volatility, which make them more advantageous than other solvents [101,102]. Researchers have successfully utilized a DES made from choline chloride and ethylene glycol to achieve a metal leaching rate of 99.3%, demonstrating the feasibility of DES in lithium-ion battery recycling and establishing the recyclability of DES [103]. Jafari et al. developed a series of nontoxic deep eutectic solvents (DES), including one ternary solvent (choline chloride (ChCl): urea: ethylene glycol (EG)) and two binary solvents (ChCl: urea and ChCl: EG), among which the ternary solvent demonstrated the best metal extraction efficiency (Li: 97%, Co: 41%, Ni: 40%, Mn: 34%). This study offers a novel, environmentally friendly, cost-effective, and safe approach to the recycling of spent lithium-ion batteries [104]. Although DES can dissolve specific metals, its leaching rates and efficiency may be lower compared to traditional acid leaching, resulting in longer recovery times. Moreover, some reagents required for preparing DES, such as choline chloride, are relatively expensive, impacting its economic viability for large-scale industrial applications. Therefore, future research should focus on process optimization and formulation improvements to increase DES application potential at an industrial scale.

Other methods: Sol-gel methods and electrochemical deposition are also employed to separate and purify metals from spent lithium-ion batteries, in addition to solvent extraction and chemical precipitation. The sol-gel method involves forming a sol from a metal precursor solution, which is then converted into a gel through hydrolysis and polycondensation reactions under specific conditions. Ultimately, the gel is dried and sintered to produce metal oxides or composite materials with excellent properties, thereby achieving metal separation and recovery. Li et al. combined the ammonia leaching and

sol-gel processes to recycle over 99% and achieved for Li, Ni, and Co [105]. Furthermore, electrochemical deposition provides energy through an external electrical medium to induce the reduction of metal ions and deposit them onto electrodes, thereby enabling the efficient recovery of key metals such as lithium, cobalt, and nickel. Owing to its straightforward process, ease of control, high product purity, and environmental cleanliness, the potential of electrochemical deposition in battery recycling is increasingly recognized. As a sustainable metal recovery technology, electrochemical deposition is expected to play a greater role in environmental protection and resource utilization in the future [106].

Table 7. Studies on separating and reclaiming high-value elements from leaching solutions.

Method	Condition	Result	Ref.
Solvent extraction	Separation factor of Co and Li(βCo/Li): 102.11 in 0.5 M HCl at 60 °C	90.5% Co, 86.2% Li	[107]
	100 g/L D2EHPA, pH = 2.7	84% Mn	[108]
	1%vol H_2O_2, D2EHPA, 90 °C, 25 g/L	99.6% Co, 98.7% Ni, 95.4% Cu, 99.5% Mn	[109]
	75 vol% D2EHPA 6h, 80 °C, 25 g/L	70% Co, 90% Li	[110]
	D2EHPA and Cyanex 272, 50 g/L, 80 °C, 4 h	99.9% $CoSO_4 \cdot H_2O$	[111]
Chemical precipitation	Na_2CO_3 for precipitating Li at 95 °C	95% Li	[112]
	Adjust pH to 5 to precipitate Mn Adjust pH to 13 to precipitate Li	93.6% Mn, 96.9% Li	[113]
	48 h, 3.5 V, 90 °C, 10g/L, different pH	Co and Li 100%	[114]
	LiOH, pH = 13, 40 °C, 45 min	Ni > 98%, and Co, Mn > 91%	[115]
	Oxidizing agent $S_2O_8^{2-}$ and the precipitant OH^-, different pH	99% Mn, 97.06% Co, 96.62% Ni	[116]
Other methods	Ammonia leaching, sol-gel	Energy density of LA achieves 924.38 $Wh \cdot kg^{-1}$	[105]
	D,L-malic acid and H_2O_2 leaching, sol-gel	Energy density achieves 602.73 $Wh \cdot kg^{-1}$	[117]
	Electrodeposited on nickel foam	Reaching 90% with excellent cycling stability after 10,000 cycles	[118]
	Density of 40 mA/m, 70 °C and pH = 2.5	99.40% Ni, 91% Co, 90.68% Mn, 85.59% Li, 89.55% Cu	[106]

4.2. Pyrometallurgy

Pyrometallurgy is the process of reducing metal oxides to metals using high temperatures. The fundamental principle involves heating the material to a sufficiently high temperature to induce chemical reactions that separate the target metals [119,120]. The pyrometallurgical recovery process for spent lithium-ion batteries can be divided into thermal reduction, smelting, and salt-assisted roasting methods, as shown in Figure 3.

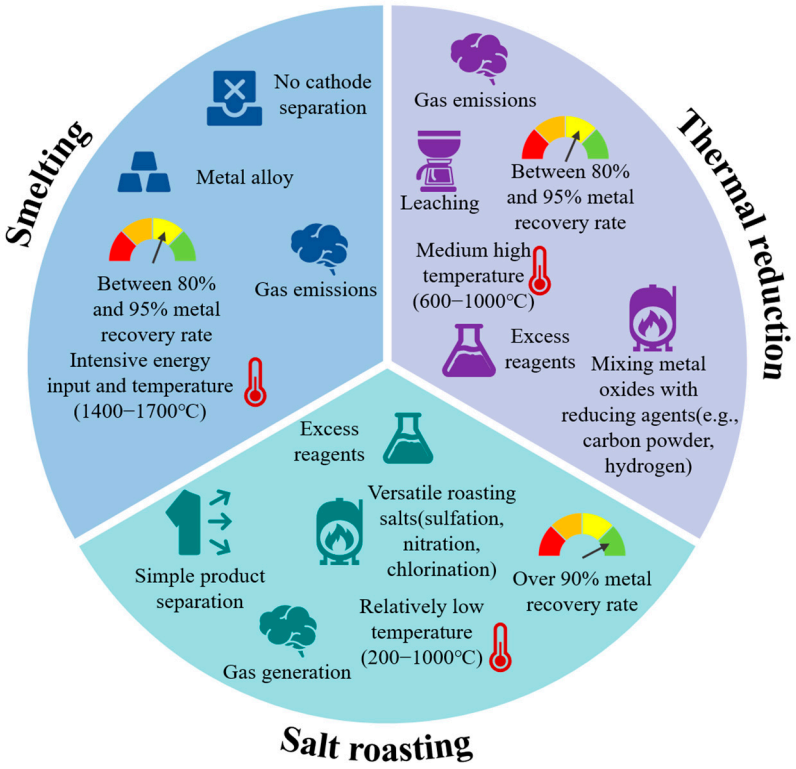


Figure 3. Characteristics of different pyrometallurgical technologies used to treat spent LIBs for the recovery of strategic metals.

Smelting: Smelting is one of the core steps in pyrometallurgy. Through high-temperature treatment, smelting melts materials and facilitates the separation of different components to achieve metal extraction. The smelting process primarily consists of two stages [50,121]: (i) The low-temperature pretreatment stage (400–600 °C), during which the cathode materials, plastics, and electrolytes of spent lithium batteries are separated and crushed into small particles. The crushed materials are then dried at low temperatures to remove moisture and organic solvents, thereby preventing the generation of unnecessary gases during smelting. Subsequently, battery materials are subjected to low-temperature heating to evaporate the electrolytes, preventing explosions due to overpressure. (ii) The high-temperature stage (800–1600 °C), where high temperatures induce redox reactions between battery waste (such as oxides in cathode materials) and additives (such as reducing agents or fluxes), thereby extracting target metals and separating impurities and slag formed from the flux. Valuable elements such as Ni, Co, Cu, and Li are present in the metal alloy and can be subsequently recovered through hydrometallurgical processes. Additionally, Li- and Mn-containing slag can be processed through hydrometallurgy or used in the cement industry [122].

Thermal reduction: Thermal reduction is a process in pyrometallurgy that converts metal oxides into metallic elements through reduction reactions at high temperatures. This technique is widely used in the recycling of LIBs, particularly for extracting valuable metals like Co, Ni, and Mn from battery waste. Prior to thermal reduction, spent lithium batteries undergo disassembly, classification, and crushing, followed by low-temperature heating to remove moisture and residual organics, which prevents excess gas generation during smelting and facilitates subsequent processes. During the thermal reduction process, active cathode materials and reducing agents are heated together; commonly used reducing agents include coke, coal, carbon monoxide, and hydrogen. These layers separate naturally at high temperatures, enabling efficient metal recovery [123]. Compared to high-temperature

smelting, thermal reduction provides advantages such as higher recovery rates of metal and lower energy consumption. Huang et al. developed a thermal reduction process using waste graphite as a reducing agent, which not only enabled the recovery of anode materials but also achieved a waste-to-waste conversion, enhancing economic benefits (Huang et al., 2019).

Salt roasting: Salt roasting is a technique that converts metal oxides into water-soluble salts by adding salt additives, facilitating the extraction of metals in subsequent leaching processes. The salt-assisted roasting process is categorized into chlorination, sulfuric acid roasting, and nitration roasting, depending on the reagents used. In lithium-ion battery recycling, salt roasting offers an advantage over other pyrometallurgical techniques due to its efficiency in metal recovery at lower temperatures, particularly suited for processing complex metal components like lithium, cobalt, and nickel, significantly enhancing metal recovery rates and efficiency. Dang et al. studied the process of chlorination roasting by adding $CaCl_2$, successfully recovering lithium in the form of LiCl [124]. Similarly, Lin et al. utilized H_2SO_4 as a sulfonating agent, achieving a selective lithium recovery rate of 99.3% and a Co_3O_4 recovery rate of 98.7% [125].

4.3. Direct Recycling

Direct recycling is an emerging lithium-ion battery recovery technology that regenerates electrode materials from spent batteries through physical or mild chemical methods while preserving their structure and chemical properties [126,127]. This approach enables the efficient recovery and reuse of lithium-ion battery materials through methods such as solvent extraction, thermal treatment, mechanical separation, and re-lithiation. Fan et al. studied the $LiNi_{1-x-y}Co_xMn_yO_2$ of spent cathode materials and proposed a targeted method for cathode recovery based on increased residual lithium compounds [128]. A satisfactory capacity was achieved for the recovered cathodes through simple sintering, enabling sustainable closed-loop recycling. Compared to traditional pyrometallurgy and hydrometallurgy, these techniques are more environmentally friendly and maximize the retention of battery materials' electrochemical performance. However, direct recycling still faces significant challenges in large-scale industrial applications, such as maintaining material purity and optimizing the recovery process costs [27,129].

4.4. Other New Methods

Electrochemically assisted leaching: Electrochemically assisted leaching employs an electric field as a reducing agent to facilitate the dissolution of metal ions in the electrolyte, converting insoluble metal ions into soluble forms and enabling metal recovery or separation through electrodeposition at the cathode [130]. The process typically consists of two fundamental steps: electrolysis and deposition. Due to its environmentally friendly nature and high selectivity, this method is widely used in battery recycling. Li et al. achieved selective lithium recovery from lithium iron phosphate (LFP) batteries through electrochemically assisted leaching, with an extraction efficiency of 99.89% [131]. Similarly, Yang et al. achieved approximately 100% lithium recovery purity from NMC batteries through direct electrochemical leaching [132]. This process incurs no additional chemical costs, thereby enhancing its economic feasibility. However, the challenges faced by this process include high costs, complex waste liquid management, and stringent operational requirements. Future research could further its potential application through technological improvements and cost reductions.

Table 8 summarizes the comparison of pyrometallurgy, direct recycling, and hydrometallurgy methods for recycling lithium-ion battery systems (LIBS).

Table 8. Comparison of main advantages and disadvantages over different LIBs recycling processes.

Process	Advantages	Disadvantages
Hydrometallurgy	It achieves high-purity metal recovery and effective separation, boasts a high recovery rate for multi-metal batteries, and operates at lower temperatures, leading to reduced energy consumption.	The process is complex, relies on large amounts of acids and bases, generates pollutants, incurs high waste treatment costs, and is slow and intricate.
Pyrometallurgy	The process is simple and suitable for large-scale processing, requires minimal waste pre-treatment for direct combustion, and operates quickly with high production efficiency.	High energy consumption; Difficult separation of complex multi-element mixtures; Emission of harmful gases, increasing environmental control costs
Direct recycling	Eco-friendly and low-pollution, it avoids chemical reagents, offers a simple operation and short processing time, consumes little energy, and retains the electrode structure for easier reuse.	Limited applicability, mainly suited to structurally simple batteries; Recovered materials may be lower in purity, with variable quality
Ref.	[130,133,134]	[135,136]

5. Conclusions

This paper provides a comprehensive summary of the current state of pre-treatment for waste lithium-ion batteries, traditional recycling methods (hydrometallurgy and pyrometallurgy), and emerging direct regeneration techniques. It examines the types of existing lithium-ion batteries, pre-treatment processes (such as discharging, dismantling, and separation), and details the recovery technologies and status of different components. Among these, pyrometallurgy and hydrometallurgy technologies are relatively established and are widely applicable in industrial production. However, pyrometallurgy requires high-temperature conditions, leading to significant energy consumption and the production of harmful gases that pollute the environment. Meanwhile, the acid-base leaching process in hydrometallurgy produces large volumes of wastewater that are challenging to treat. Although bioleaching mitigates this issue, its leaching rate remains low and requires further improvement. Consequently, several novel non-toxic methods have begun to emerge and advance. Technologies such as electrochemical methods and deep eutectic solvents not only meet the requirements for low energy consumption, low emissions, and high efficiency but also enhance the options for waste lithium-ion battery recycling. However, these innovations bring new challenges, necessitating the optimization of reaction conditions and the identification of more environmentally friendly and economical reagents. Looking ahead, the recycling of waste lithium-ion batteries will remain a critical issue in the field of environmental protection. Exploring new methods is essential to draw attention and expand prospects in the battery recycling field.

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