

# One-Step Recovery and Regeneration Technology for Discarded Lithium-Ion Battery Cathode Materials

Hong Chen, Lu-Kang Zhao, Xuan-Chen Wang, Xuan-Wen Gao,\* Qing-Song Lai, Zhao-Meng Liu,\* Dong-Run Yang, and Wen-Bin Luo\*

Fossil fuels have been instrumental in propelling the rapid advancement of human civilization. However, the carbon emissions resulting from their combustion have given rise to environmental challenges. The notable increase in the sales of new energy vehicles has positively contributed to lowering fuel consumption and aligning with national objectives for green and sustainable development. Nevertheless, the growing number of new energy vehicles presents challenges for the handling and recycling of lithium-ion batteries (LIBs). Improper handling can result in environmental pollution. Scholars worldwide are

actively exploring effective eco-friendly management solutions for managing discarded LIBs, with a particular concern about recycling metal materials. This review thoroughly examines the failure mechanisms of LIBs cathode materials and traditional recycling methods, assesses one-step recovery and regeneration technologies, and explores the challenges, technologies, and future prospects of key material recycling in the next generation of batteries. The comprehensive insights provided in this review aim to contribute to the industrialization and scaling up of future power battery recycling technologies.

## 1. Introduction

Fossil fuels, such as coal and petroleum, have been instrumental in the rapid progress of human civilization.<sup>[1,2]</sup> However, the combustion of these energy sources has led to severe environmental challenges, including the greenhouse effect due to carbon emissions.<sup>[3,4]</sup> China has proposed the goal of achieving “peak carbon emissions and carbon neutrality” by 2060 to address this issue. Against the backdrop of this global green transformation, the sales of new energy vehicles have experienced explosive growth, making a positive contribution to reducing fuel consumption and aligning with the national goals of green and sustainable development.<sup>[5,6]</sup>

However, as the first generation of electric vehicles equipped with lithium-ion battery (LIB) packs is gradually being phased out and the number of new energy vehicles continues to increase, the recycling and utilization of LIBs has become an increasingly prominent issue.<sup>[7–10]</sup> Waste LIBs contain a range of metal resources, including lithium, cobalt, nickel, manganese, iron, copper, and aluminum.<sup>[11,12]</sup> If these waste batteries are not properly treated and are directly discarded, they can have a serious impact on the environment.<sup>[13–15]</sup> These metals may leach into the soil and groundwater, leading to soil and water pollution.<sup>[16]</sup> Additionally, the electrolyte in the batteries is prone to releasing toxic gases when reacting with water, further exacerbating the

risk of environmental pollution.<sup>[17]</sup> Therefore, the presence of these metal pollutants not only affects the ecosystem but also poses a threat to human health.<sup>[18]</sup>

Faced with these environmental and safety challenges, effective measures must be taken to environmentally process waste LIBs and recover the resources within them, which is of paramount importance.<sup>[19,20]</sup> This has garnered widespread attention from scholars worldwide, becoming a focal point of research. In particular, the reprocessing and reutilization of metal materials from waste LIBs have been given significant emphasis.<sup>[21–23]</sup>

Thus, this review investigates and elaborates on the failure mechanisms of LIB cathode materials and traditional recycling techniques. In addition, it summarizes and evaluates integrated regeneration technologies. Finally, it introduces and analyzes the challenges, technologies, and future prospects of the next-generation battery’s key material recycling (Figure 1). Through this review, we aim to systematically introduce the latest developments in this field to readers and, more importantly, provide insights for the industrialization and scale-up of future power battery recycling technologies.

## 2. The Failure Mechanisms of Positive Electrode Materials

The failure mechanism of LIBs has always been a focal point for researchers because it directly impacts the capacity and lifespan of LIBs.<sup>[24]</sup> Battery degradation typically occurs in multiple components, including anode, cathode, electrolyte, separator, and current collector.<sup>[25]</sup> Among these, degradation issues with the cathode material include cation mixing, side reactions, cracking, and oxygen release, which directly affect the battery’s capacity and safety. The most significant of these is cathode material decay,

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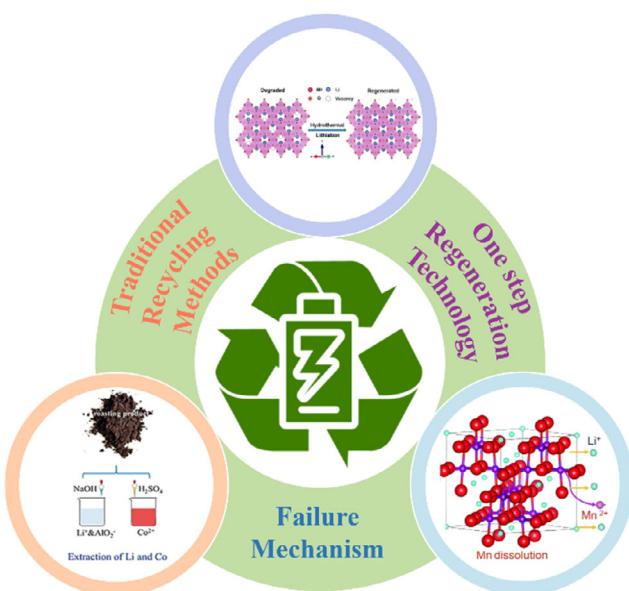
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**Figure 1.** Main body of the article. Reproduced with permission.<sup>[94–96]</sup> Copyright 2016, Wiley; Copyright 2019, American Chemical Society; and Copyright 2020, American Chemical Society.

which can lead to decreased battery performance, reduced energy storage capacity, and increased safety risks. Therefore, it is crucial to have a thorough understanding of the material depletion mechanism of the cathode to improve the performance and lifespan of LIBs.

The degradation of LIBs typically occurs in the cathode, anode, electrolyte, separator, and current collector. Issues related to the degradation of the cathode material, such as cation mixing, side reactions, cracking, and oxygen release, have a direct impact on the capacity and safety of the battery.

## 2.1. Cation Mixing

When using nickel-rich layered oxide as the cathode material, increasing the nickel (Ni) content enhances its capacity. This is



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due to the three oxidation states of nickel, namely +2, +3, and +4, allowing for the adjustment of oxidation states to balance charge variations and achieve higher capacity.<sup>[26]</sup> However, as the nickel content is increased, the cycling stability of the cathode material is significantly reduced. This is primarily because the ionic radius of  $\text{Ni}^{2+}$  (0.069 nm) is close to that of  $\text{Li}^+$  (0.076 nm), making it easy for Ni to occupy Li positions, leading to Li/Ni mixing, and causing changes in the crystal structure, accelerating the decay of material capacity (Figure 2A).<sup>[27]</sup> Additionally, as Ni occupies some positions in the Li layers, it hinders the migration rate of  $\text{Li}^+$  in the layered material, resulting in an increase in insertion impedance, thus, affecting rate performance. When all  $\text{Li}^+$  positions are fully occupied by  $\text{Ni}^{2+}$ , the cathode material undergoes a structural transformation from the layered phase ( $\text{R}3\text{m}$ ) to the spinel phase ( $\text{Fd}3\text{m}$ ) and rock salt phase ( $\text{Fm}3\text{m}$ ). This transformation further hinders  $\text{Li}^+$  diffusion and results in reduced thermal stability.<sup>[28]</sup> The mixing of Li/Ni takes place not just in the course of material synthesis but also throughout the charging and discharging processes, exerting a substantial impact on the electrochemical characteristics, including initial capacity, reversible capacity, and cycling performance of the material.

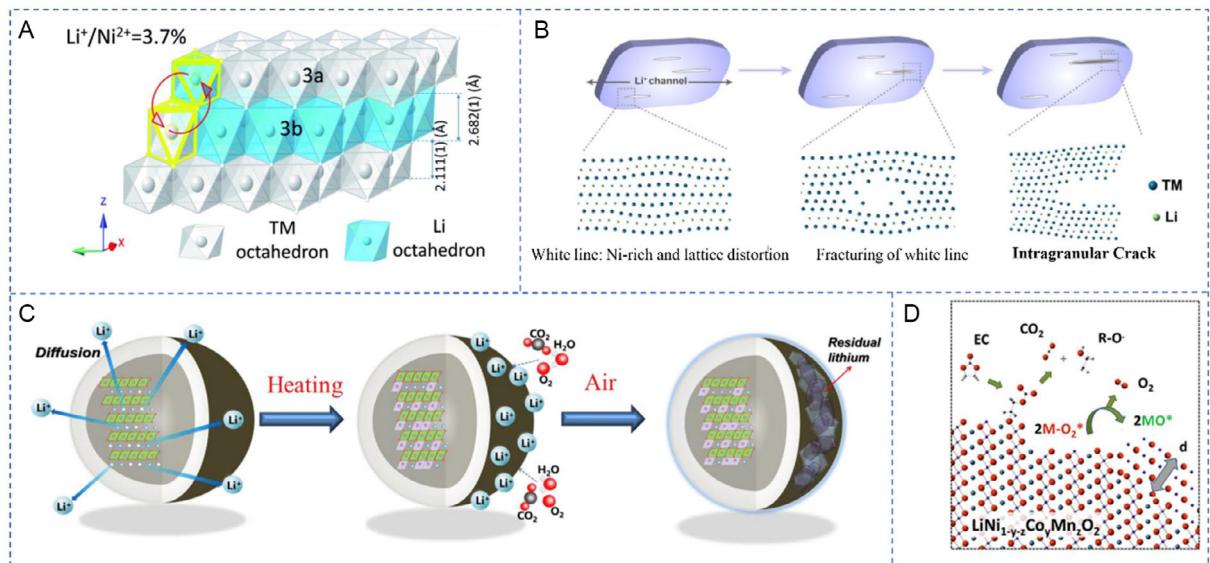
## 2.2. Generation of Microcracks

The main cause of electrode material instability is the generation of cracks in the crystal structure. Cracks can be classified into two types:<sup>[29]</sup> intergranular cracks located at grain boundaries and intragranular cracks within the grains.

The formation of intragranular cracks is primarily attributed to the uneven distribution of  $\text{Li}^+$ . Li/Ni mixing leads to a lack of Li in the rock salt layer, hindering the diffusion of  $\text{Li}^+$  and causing uneven distribution of  $\text{Li}^+$  in the anode structure. Due to the uneven distribution of Li, internal stress affects the particles. Figure 2B shows the weakening of the O electrostatic shielding effect between adjacent layered structures due to the removal of  $\text{Li}^+$ . This removal causes an increase in the Li layer spacing and the lattice parameter  $c$ , which ultimately leads to the collapse of the entire crystalline structure and the evolution of cracks.<sup>[30,31]</sup>



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**Figure 2.** A) Schematic diagram of  $\text{Ni}^{2+}/\text{Li}^+$  cation mixing. Reproduced with permission.<sup>[27]</sup> Copyright 2014, Royal Society of Chemistry. B) The mechanism underlying the formation and development of intragranular cracks. Reproduced with permission.<sup>[31]</sup> Copyright 2020, Elsevier. C) Illustration of the formation process of  $\text{LiOH}$  and  $\text{Li}_2\text{CO}_3$ . Reproduced with permission.<sup>[33]</sup> Copyright 2016, Elsevier. D) Schematic representation of key reactions leading to surface reconstruction, as well as the generation of  $\text{CO}_2$  and  $\text{O}_2$ . Reproduced with permission.<sup>[36]</sup> Copyright 2017, American Chemical Society.

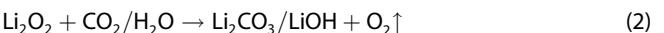
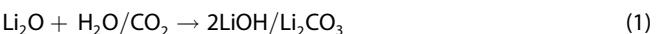
Over the course of the cycle, the layered phase undergoes repeated volume expansion/contraction due to  $\text{Li}^+$  insertion/extraction, while the adjacent rock salt phase maintains a constant crystal volume due to a lack of electrochemical activity. This phase mismatch leads to an increase in internal stress, ultimately causing crack formation. Simultaneously, the release of oxygen and oxygen vacancies accelerates the generation of intragranular cracks.

The transition from H2 to H3 phase in the material also results in the development of intergranular cracks. During the phase transition, the lattice parameters drastically decrease, causing the collapse of the battery structure and the generation of intergranular cracks. During charge and discharge processes, the repeated insertion/extraction of  $\text{Li}^+$  causes isotropic changes in the primary particles, leading to the accumulation of local stress at grain boundaries, ultimately resulting in the formation of intergranular cracks. Additionally, the distortion of secondary particles during long-term cycling and lattice contraction causes the formation of expansive cracks within the particles. With an increase in nickel content, the cracks gradually enlarge.<sup>[32]</sup> These cracks reduce the interparticle connection, resulting in a decrease in secondary particle conductivity and an increase in resistance. The ingress of electrolyte into the cracks also facilitates side reactions and electrode corrosion. Consequently, the development of cracks has a direct impact on the performance and lifetime of LIBs.

### 2.3. Interfacial Side Reactions

The surface/interface interaction between the cathode material and the electrolyte in LIBs often triggers a series of unavoidable side reactions. This is due to the presence of an excess of lithium oxides, such as  $\text{LiO}_2$  and  $\text{Li}_2\text{O}_2$ , on the surface of the cathode material during the preparation of nickel-rich layered oxide positive electrode materials, forming a series of by-products

collectively referred to as residual lithium (Figure 2C).<sup>[33]</sup> The cathode material is highly sensitive to  $\text{O}_2$  and  $\text{H}_2\text{O}$  in the air, leading to the formation of by-products, such as  $\text{Li}_2\text{CO}_3$  and  $\text{LiOH}$ , which not only lack electrochemical activity but also consume a significant amount of  $\text{Li}^+$ , resulting in a reduction in the cathode material's capacity. In particular,  $\text{LiOH}$  reacts with  $\text{LiPF}_6$ , consuming a large amount of  $\text{Li}^+$  in the electrolyte and generating HF, thereby corroding the battery structure and causing instability in the entire cathode. Additionally,  $\text{Li}_2\text{CO}_3$  significantly hinders the transport of  $\text{Li}^+$  on the cathode surface, leading to irreversible capacity decay during the cycling process. It is also noteworthy that  $\text{Li}_2\text{CO}_3$  reacts with the O atoms overflowing from the  $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$  (NCM) material, generating  $\text{CO}_2$  gas, causing battery expansion and further jeopardizing the safety of the battery structure.<sup>[34,35]</sup> The presence of these by-products from side reactions also results in poor conductivity and increased resistance at the interface. The overall reaction process is as follows, mainly involving the following two formulas



Reaction 1 and Reaction 2 jointly constitute the entire reaction process.

### 2.4. Oxygen Release and Excessive Metal Dissolution

For high-nickel positive electrode materials, the occurrence of oxygen release and the dissolution of transition metals are inevitable issues during the battery operation. The enhanced capacity of nickel-rich materials is ascribed to the fluctuation in nickel oxidation states. Simultaneously, a substantial quantity of  $\text{Li}^+$  is extracted from the layered structure. To maintain the charge balance of

the entire electrode material, oxygen in the lattice is released, leading to surface reconstruction. The released oxygen reacts further with the electrolyte, initiating redox reactions that result in battery failure (Figure 2D).<sup>[36]</sup> The surface of positive electrode particles undergoes severe corrosion due to by-products from side reactions, such as HF. This corrosion, coupled with the dissolution of metal elements like Mn, results in the depletion of active materials. Among the transition metal elements, Mn is one of the most readily dissolved elements.<sup>[37]</sup> Gummow et al. reported a dissolution model for Mn in acidic aqueous media, represented by  $\text{LiMn}_2\text{O}_4 + 4 \text{H}^+ \rightarrow 2\text{Li}^+ + \text{Mn}^{2+} + 3\lambda\text{-MnO}_2 + 2\text{H}_2\text{O}$ .<sup>[38]</sup> During the dissolution process,  $\text{Mn}^{2+}$  reacts vigorously with other carbonate solvent molecules and electrolyte anions, resulting in the formation of Mn complexes. These complexes precipitate onto the positive electrode surface as by-products, which leads to elevated interface impedance, and diminished cycling stability and rate performance of the positive electrode.

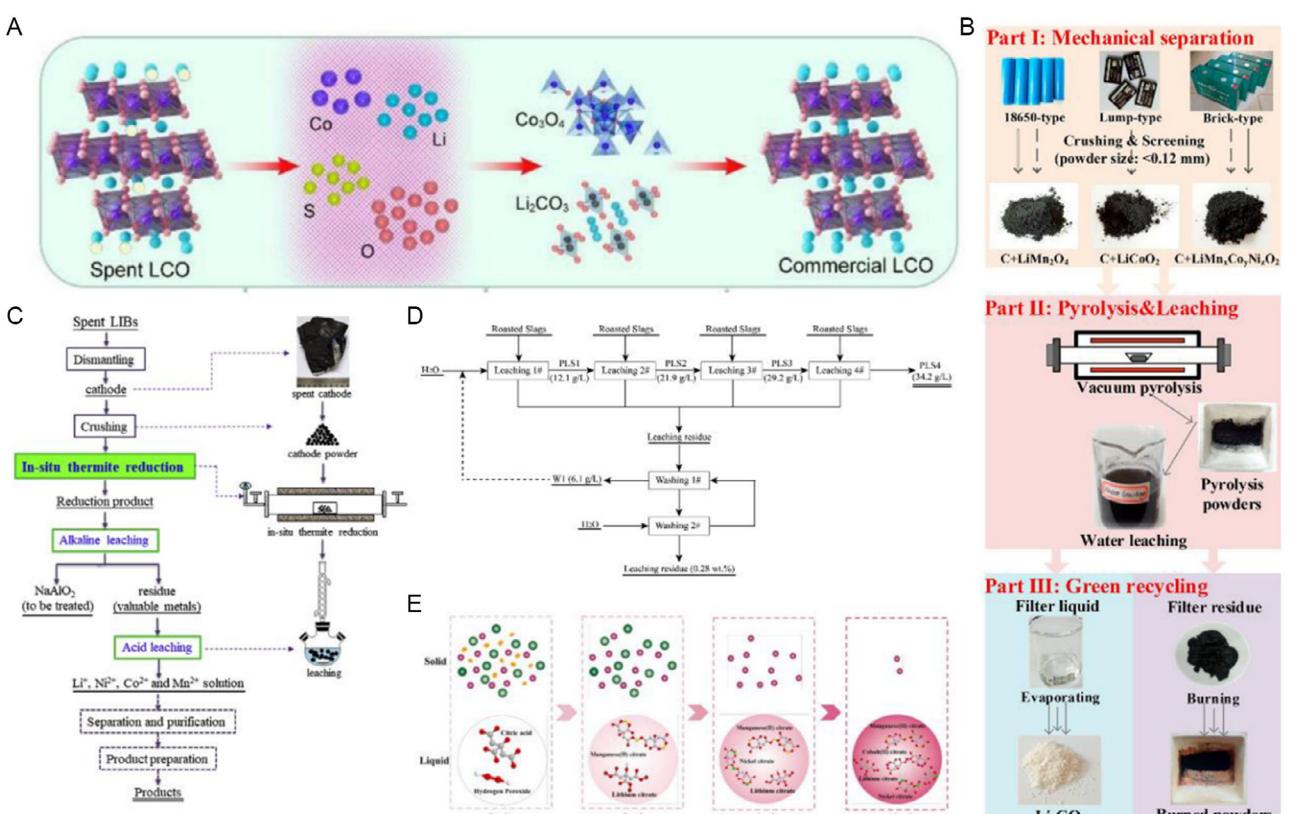
### **3. Traditional Recycling Methods**

### 3.1. Traditional Pyrometallurgical Processing Methods

Traditional pyrometallurgical methods have become the most widely used approach for recycling LIBs owing to their effortless

operation and elevated recovery rates.<sup>[39]</sup> Nevertheless, to form alloys in the battery materials, pyrometallurgy typically requires extremely high temperatures. Consequently, pyrometallurgical processes suffer from significant drawbacks, such as substantial losses of valuable metals in the slag and high energy consumption.

The salt-assisted roasting (SAR) technology is effective in minimizing the calcination temperature in contrast to conventional pyrometallurgy, transforming metal oxides into soluble products in water. This allows for the rapid and easy destruction of the structure of positive electrode materials at lower temperatures, greatly improving overall recovery efficiency. Depending on the solvent used, SAR can be categorized into sulfuric acid roasting,<sup>[40,41]</sup> chloride roasting,<sup>[42,43]</sup> and nitrate roasting.<sup>[44]</sup> As depicted in Figure 3A, Wang et al. proposed an environmentally friendly recovery method in which the inclusion of ammonium sulfate resulted in a decreased roasting temperature, below 400 °C, thereby significantly reducing energy consumption. By using inexpensive reagents and water as a leaching agent, the recovery process is expected to yield significant benefits, with an estimated value of \$6.94 kg<sup>-1</sup> of battery.<sup>[45]</sup> Ma et al. presented a low-temperature chlorination pyrolysis process for the selective recovery and recycling of valuable metals. Initially, spent-NCM (s-NCM) positive electrode materials were thermally treated with NH<sub>4</sub>Cl as a chlorinating agent. The valuable metals (Ni, Co, Mn, Li) were converted into



**Figure 3.** A) Diagram of proposed recovery processing using ammonium sulfate assisted roasting. Reproduced with permission.<sup>[45]</sup> Copyright 2022, Elsevier. B) Integrated process of in situ roasting for recovering Li<sub>2</sub>CO<sub>3</sub> from used LIBs. Reproduced with permission.<sup>[49]</sup> Copyright 2017, American Chemical Society. C) In situ aluminum thermal agent reduction process for recycling cathodes of spent LIBs. Reproduced with permission.<sup>[48]</sup> Copyright 2020, Elsevier. D) Process flow diagram of cross-current leaching in nitric acid roasting for LIB raw materials. Reproduced with permission.<sup>[47]</sup> Copyright 2019, Elsevier. E) Schematic illustration of the leaching mechanism proposed based on ammonium sulfate leaching. Reproduced with permission.<sup>[66]</sup> Copyright 2017, Elsevier.

soluble chlorides by the liberation of HCl and NH<sub>3</sub> from NH<sub>4</sub>Cl. By subjecting the material to low-temperature chlorination pyrolysis at 300 °C for 30 min with a Cl/s-NCM mass ratio of 3:1, over 99.9% of the valuable metals could be reclaimed through simple water leaching.<sup>[46]</sup> As shown in Figure 3D, Peng et al. introduced a recovery process involving nitration and selective roasting. The metallic components of LIB waste were first converted into corresponding nitrates, which were then broken down into insoluble oxides, except for lithium nitrate, which could be extracted through leaching. Selective roasting at 250 °C resulted in a peak lithium extraction rate of 93%, while the extraction rate of other metals (such as cobalt, nickel, copper, and so on) remained below 0.1%. Implementing this innovative process can significantly enhance the lithium recovery efficiency of waste LIBs, achieving an overall lithium recovery rate of 90%.<sup>[47]</sup>

In situ roasting recovery technology is the conversion of waste into valuable products using the components of the waste positive electrode material itself without adding reagents or components, embodying the concept of “waste + waste = resource”. In contrast to conventional carbothermal reduction roasting methods, the in situ roasting recovery technology eliminates the requirement for supplementary carbon materials (such as graphite, wood charcoal, coke, and so on) as reducing agents. This substantial cost-saving approach also simplifies separation procedures. For example, as shown in Figure 3C, Wang et al. utilized the aluminum preform collector as an in situ reducing agent for aluminum thermal reduction, converting the precious metals in LiNi<sub>x</sub>Co<sub>y</sub>Mn<sub>z</sub>O<sub>2</sub> cathodes into LiAlO<sub>2</sub>, Li<sub>2</sub>O, NiO, CoO, and MnO. Subsequently, aluminum iron ore reduction products were effectively leached to remove Al using alkali leaching without significant explosive H<sub>2</sub> release. Subsequently, valuable metals Li, Ni, Co, and Mn were effectively extracted into an H<sub>2</sub>SO<sub>4</sub> solution, achieving leaching efficiencies of 99.78, 98.62, 99.29, and 99.91%, respectively.<sup>[48]</sup> As shown in Figure 3B, Xiao et al. proposed a new method for recovering lithium elements from waste LIBs positive electrode materials as Li<sub>2</sub>CO<sub>3</sub>. The positive electrode material obtained from pyrolysis pretreatment under closed vacuum conditions disrupts the oxygen framework, releasing Li from the crystal structure of lithium transition metal oxides in the form of Li<sub>2</sub>CO<sub>3</sub>. A lithium recovery rate of 81.90% and Li<sub>2</sub>CO<sub>3</sub> purity of 99.7% was achieved.<sup>[49]</sup> Despite the simplicity and maturity of pyrometallurgical processing methods, challenges such as high energy consumption, severe environmental pollution, and difficulties in recovering other components of the battery still exist.

### 3.2. Traditional Hydrometallurgical Processing Methods

Wet metallurgy refers to an approach employing low-temperature leaching and separation processes for the recovery of valuable metals from spent LIBs cathode materials.<sup>[50]</sup> The resulting filtrate, which contains a variety of valuable metals, is further purified by various methods. Ultimately, this refined process leads to the production of new cathode materials or high-value-added products, maximizing the overall recovery rate.<sup>[51]</sup> Leaching is a crucial step in wet metallurgy, with conventional leaching methods including acidic leaching and alkaline leaching.

Indeed, acid leaching can be categorized into two main types based on the solvent used: inorganic acid leaching and organic acid leaching. Compared to alkaline leaching, acidic leaching can maximally dissolve metals from the cathode materials. Therefore, acidic leaching remains the most widely researched method for metal extraction. Common inorganic acids include hydrochloric acid (HCl), nitric acid (HNO<sub>3</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), and phosphoric acid (H<sub>3</sub>PO<sub>4</sub>).<sup>[52–56]</sup> Due to the presence of high valence metals (such as Co<sup>3+</sup> and Ni<sup>4+</sup>) in the cathode materials, a reducing agent is typically introduced to convert them to their respective divalent oxides, thereby improving the solubility of the metals. However, most organic acids are strong acids, and their use can lead to the formation of a lot of acidic and toxic gases, causing environmental pollution and health hazards. Therefore, the research focus has shifted to organic acids. Organic acid solvents currently in use include, malic acid, maleic acid, lactic acid, acetic acid, oxalic acid, formic acid, citric acid,<sup>[57–60]</sup> etc. Due to the unique chelating properties of organic acids, they are capable of extracting valuable metals from leachate and regenerating into new cathode materials. This minimizes the need for additional recovery processing steps. For example, Li et al. proposed a process for recycling waste mixed with cathode materials (LiMn<sub>2</sub>O<sub>4</sub>, LiCoO<sub>2</sub>, and Li Ni<sub>1/3</sub>Co<sub>1/3</sub>Mn<sub>1/3</sub>O<sub>2</sub>) using the sol-gel method to resynthesize new LiNi<sub>1/3</sub>Co<sub>1/3</sub>Mn<sub>1/3</sub>O<sub>2</sub> materials. Comparative studies have shown that the electrochemical performance of the resynthesized material (NCM-spent) is superior to that of the directly synthesized material (NCM-syn). The initial discharge capacity of NCM-spent at 0.2C was 152.8 mA h g<sup>-1</sup>, which exceeded that of NCM-syn (149.8 mA h g<sup>-1</sup>). The improved performance of NCM-spent can be attributed to the presence of trace aluminum doping.<sup>[61]</sup>

Alkaline solutions use the reaction between hydroxide ions and metals to recover cathode materials. The advantage of alkaline leaching is the ability to selectively leach, avoiding complex separations. Since alkaline solutions can dissolve Al while Co, Ni, Mn, etc., cannot, NaOH solution can be employed to remove Al, and amine solutions are typically used for materials containing Co, Ni, Mn, Li, and other metals. Typical reagents employed in ammonia dissolution include ammonia (NH<sub>3</sub>), ammonium sulfate [(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>], ammonium sulfite [(NH<sub>4</sub>)<sub>2</sub>SO<sub>3</sub>], ammonium carbonate [(NH<sub>4</sub>)<sub>2</sub>CO<sub>3</sub>], ammonium hydroxide [(NH<sub>4</sub>)OH], and ammonium oxalate [(NH<sub>4</sub>)<sub>2</sub>C<sub>2</sub>O<sub>4</sub>].<sup>[62–65]</sup> Ammonium ions form ammonia complexes with metal ions, significantly increasing the metal leaching rate for further recovery. For example, as illustrated in Figure 3E, Zheng et al. developed a thorough hydrometallurgical method to exclusively recover the precious metals (Ni, Co, and Li) contained in the cathode waste of depleted LIBs. Employing ammonium sulfate as the leaching solution and sodium sulfite as the reducing agent, the overall selectivity for Ni, Co, and Li in the initial leachate surpassed 98.6%, whereas, the total selectivity for Mn amounted to merely 1.36%. The investigation found that Mn primarily transitioned into the solution as Mn<sup>4+</sup>, which was then reduced to Mn<sup>2+</sup>, and subsequently precipitated in the form of (NH<sub>4</sub>)<sub>2</sub>Mn(SO<sub>3</sub>)<sub>2</sub>·H<sub>2</sub>O in the residue. Meanwhile, Ni, Co, and Li were generally leached and retained in the dissolution as metallic ions or amino composites.<sup>[66]</sup>

In summary, the mentioned hydrometallurgical processes significantly improve the pollution issues faced by traditional pyrometallurgy, representing innovative approaches worthy of further exploration. However, both traditional hydrometallurgy and pyrometallurgy have high energy consumption, lengthy processes, and challenges for large-scale applications. Hence, it is crucial to explore comprehensive recovery and regeneration technologies for spent LIB cathode materials that align with the criteria of energy conservation, efficiency, simplicity, and environmental friendliness.

#### 4. One-Step Regeneration Technology

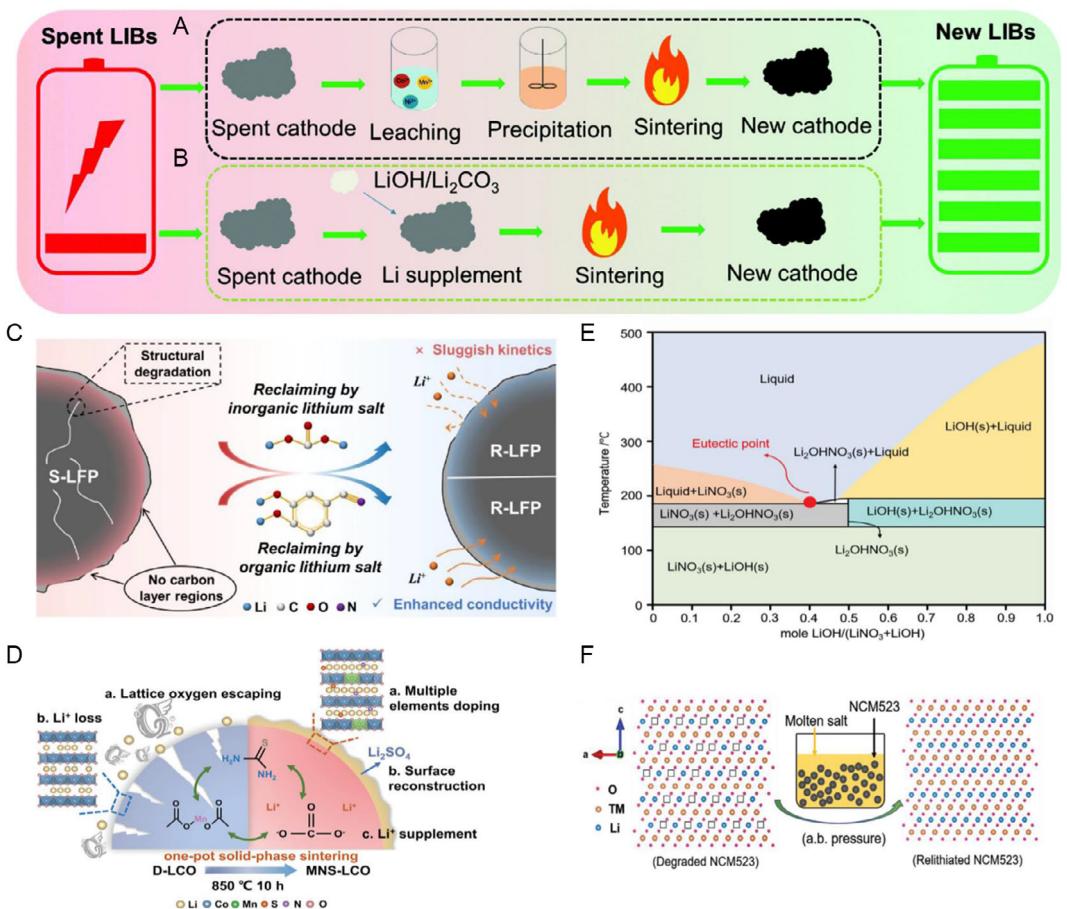
The closed-loop pathway for the integrated regeneration of positive electrode materials is illustrated in Figure 4A, which primarily utilizes coprecipitation and sol-gel methods for the regeneration process. Compared to traditional regeneration methods, this recycling pathway simplifies processes, reduces the use of chemical reagents, minimizes secondary pollution, and greatly enhances the overall recovery performance of used batteries.

#### 4.1. Sol-Gel Method

The first regeneration method is the sol-gel method. Depending on the different leaching agents, the most appropriate regeneration method is selected. After leaching with organic acid, the sol-gel process is commonly applied to directly regenerate new positive electrode materials. This process utilizes the acidic and chelating properties of the organic acid. Examples of such organic acids are malic acid,<sup>[67]</sup> lactic acid,<sup>[68]</sup> and citric acid.<sup>[69,70]</sup> For example, Zhang et al. used D, L-malic acid and H<sub>2</sub>O<sub>2</sub> to extract spent lithium cobalt oxide, and through the sol-gel method, they resynthesized LiCo<sub>1/3</sub>Ni<sub>1/3-y</sub>Al<sub>y</sub>Mn<sub>1/3</sub>O<sub>2</sub> ( $y = 0.02, 0.04$ ). The material obtained exhibited a typical layered structure. After 100 cycles at 0.2C current density, the reversible discharge capacity remained at 148.1 mAh g<sup>-1</sup>, surpassing that of LiCo<sub>1/3</sub>Ni<sub>1/3</sub>Mn<sub>1/3</sub>O<sub>2</sub>.<sup>[67]</sup>

#### 4.2. Coprecipitation Method

The coprecipitation method is the second regeneration method.<sup>[71–73]</sup> The process consists of several steps: first, impurities are removed from various metal salts. Then, the components in the metal leaching solution are adjusted to the appropriate ratio.



**Figure 4.** New cathode material synthesis in a closed loop: A) regeneration and B) direct recycling. (A,B) Reproduced with permission.<sup>[97]</sup> Copyright 2022, Royal Society of Chemistry. C) Diagram of S-LFP regeneration mechanism using inorganic and organic lithium salts. Reproduced with permission.<sup>[92]</sup> Copyright 2023, Springer Nature. D) Schematic representation of the upcycling process of D-LCO cathodes. Reproduced with permission.<sup>[93]</sup> Copyright 2023, Wiley. E) Equilibrium phases of LiNO<sub>3</sub> and LiOH, sourced from the FACT salt database.<sup>[91]</sup> F) Diagram of the relithification process for Li composition recovery via the eutectic molten salt approach. (E,F) Reproduced with permission.<sup>[91]</sup> Copyright 2019, Wiley.

Next, a coprecipitating agent is added to produce the precursor of the positive electrode material. Finally, the new positive electrode material is regenerated by solid-phase sintering the mixture of the precursor and lithium salt. In a practical application, the effectiveness of this method is also influenced by the type of leaching agent used. Organic acids can form complexes with metal ions, altering their properties and hindering their use in regenerating new positive electrode material precursors through coprecipitation. For this reason, the sol-gel process is often used for metal ion regeneration after organic acidic leaches. In contrast, the regeneration system for inorganic acid leaching adopts the coprecipitation regeneration method without worrying about affecting the precipitation performance of transition metals.<sup>[74]</sup> For example, Yang et al. used sulfuric acid and hydrogen peroxide to successfully regenerate manganese-rich  $0.2\text{Li}_2\text{MnO}_3\text{--}0.8\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  positive electrode material from the leachate of mixed waste positive electrode materials ( $\text{LiMn}_2\text{O}_4$ ,  $\text{LiCoO}_2$ , and  $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$  positive electrode materials).<sup>[72]</sup> The experimental results showed that under the conditions of  $\text{pH} = 10.5$  and  $0.5 \text{ M NH}_3$ , spherical precursors with good particle size and crystallinity were obtained. The precursor was mixed with lithium hydroxide and calcined to produce manganese-rich  $0.2\text{Li}_2\text{MnO}_3\text{--}0.8\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  positive electrode material. The regenerated positive electrode material retained the morphological characteristics of the precursor and exhibited a uniform particle size distribution. The regenerated positive electrode material had a layered structure with low cation mixing. The regenerated positive electrode material exhibited good electrochemical performance, with discharge capacities of 248.3, 196.4, and  $167.0 \text{ mAh g}^{-1}$  after 50 cycles at constant current densities of 0.1, 0.5, and 1C, respectively, and capacity retention rates of about 88, 85, and 80%, respectively. He et al. used spent LIBs as raw materials and synthesized spherical  $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  positive electrode material by the carbonate coprecipitation method.<sup>[73]</sup> The regenerated  $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  consisted of spherical particles similar to the  $\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{CO}_3$  precursor, but with a narrower particle size distribution. In addition, it had a well-ordered layered structure and a low degree of cation mixing. The regenerated  $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  exhibited excellent electrochemical performance (discharge capacity of  $163.5 \text{ mAh g}^{-1}$  at 0.1C, discharge capacity of  $135.1 \text{ mAh g}^{-1}$  at 1C, and capacity retention rate of 94.1% after 50 cycles). Even at a high rate of 5C, it could provide a high capacity of  $112.6 \text{ mAh g}^{-1}$ .

In summary, each regeneration method possesses distinct advantages and inherent limitations. Going forward, there is a continual need for research and development of innovative regeneration strategies to optimize the recovery of spent LIBs, with the ultimate goal of realizing efficient, environmentally friendly, and convenient recycling.

#### 4.3. The Resynthesis of other High-Value-Added Products

The recycling of spent LIBs includes not only the regeneration of new cathode materials, but also the regeneration of other high-value products, such as metal-organic frameworks (MOFs), nanostructured  $\text{Co}_3\text{O}_4$  anodes,<sup>[75]</sup> supercapacitors,<sup>[76,77]</sup> magnetic  $\text{CoFe}_2\text{O}_4$ ,<sup>[78]</sup> catalysts, and more.<sup>[79]</sup>

Mao et al. employed leachate from spent LIBs as a cobalt source and successfully synthesized  $\text{Co}_3\text{O}_4$  hollow microspheres through a solvothermal method.<sup>[76]</sup> The research findings revealed that the  $\text{Co}_3\text{O}_4$  hollow microspheres (with a diameter less than  $1 \mu\text{m}$ ) possessed numerous mesopores, advantageous for the fabrication of highly efficient supercapacitor electrodes. Simultaneously, the  $\text{Co}_3\text{O}_4$  hollow microspheres exhibited elevated specific capacitance and excellent stability. Hu et al. recovered cobalt oxalate from spent LIBs using alkaline leaching, acid leaching followed by reduction, and chemical precipitation.<sup>[75]</sup> Using the recovered cobalt oxalate as a precursor, they synthesized nanostructured  $\text{Co}_3\text{O}_4$  cathode materials by the sol-gel method. The influence of the molar ratio of  $\text{Co}^{2+}$  to citric acid and the calcination temperature on the  $\text{Co}_3\text{O}_4$  structure and electrochemical performance was investigated.

Jiao et al. developed an innovative approach to upcycling waste LIBs by integrating Co/Fe species with wood-derived carbon to construct a CoFe/C single-atom catalyst.<sup>[80]</sup> Their study revealed that during the annealing process, the electrostatic interactions between  $\text{Co}^{3+}/\text{Fe}^{3+}$  and lignin hydroxyl groups facilitated the homogeneous dispersion of CoFe nanoparticles within the carbon matrix. Advanced atomic-scale characterization further demonstrated that Fe atoms were embedded in the Co lattice as single atoms, inducing electron redistribution and activating a Pt-like dissociation mechanism. As a result, the catalyst exhibited a high power density of  $199.2 \text{ mW cm}^{-2}$  and maintained stable operation for 350 h in zinc-air batteries. Furthermore, its flexible configuration successfully powered LEDs and portable electronic devices, highlighting its potential for practical applications.

Cheng et al. proposed a high-efficiency strategy for directly converting spent NMC cathodes into NiMnCo-activated carbon core-shell catalysts via acid leaching combined with radiation heating.<sup>[81]</sup> The resulting catalyst consists of a face-centered cubic Ni core and a spinel  $\text{NiMnCoO}_4$  shell, where interfacial electronic reconstruction significantly lowers the overpotentials for both the oxygen reduction and oxygen evolution reactions. When employed as a zinc-air batteries cathode, this material delivered an impressive power density of  $187.7 \text{ mW cm}^{-2}$  and demonstrated exceptional cycling stability, maintaining operation for 200 h at a current density of  $10 \text{ mA cm}^{-2}$ , with an initial voltage gap as low as 0.72 V.

Zhu et al. developed a high-temperature shock method to transform waste  $\text{LiCoO}_2/\text{LiMn}_2\text{O}_4$  cathodes into a Co-MnO bifunctional catalyst.<sup>[82]</sup> The strong synergistic effect between Co and Mn facilitated efficient polysulfide anchoring and catalytic conversion, significantly improving lithium-sulfur battery performance. Even with a catalyst loading of less than 0.5 wt%, the lithium-sulfur battery retained a high specific capacity of  $707 \text{ mAh g}^{-1}$  at a 4C, with a remarkably low capacity decay rate of only 0.058% per cycle over 400 cycles.

Moreover, the cathode powder from spent LIBs can be utilized for the degradation of industrial pollutants, exemplifying the concept of "using waste to treat waste".<sup>[79]</sup> Utilizing the excellent dielectric properties of waste lithium cobalt oxide powder, it can effectively remove sulfur from wastewater.<sup>[83]</sup> Recycling methods for electronic waste and industrial waste deserve

thorough study and promotion to achieve maximum economic and environmental benefits.

#### 4.4. The Direct Regeneration Process

The direct regeneration process is an eco-friendly recycling method that entails the direct retrieval and recovery of positive electrode materials from LIBs without causing harm to their original structure.<sup>[84]</sup> This approach accomplishes the revitalization of positive electrode active materials by reinstating the structure of the positive electrode active material and replenishing the lost active lithium ions during the cycling process, eliminating the necessity for leaching treatment. The advantages of direct regeneration lie in its simplification of complex recovery steps, significant reduction in recovery costs, and substantial reduction in the emission of pollutants, such as exhaust gases and wastewater, during the recovery process. Because this direct recovery method can efficiently recover the original lithium and restore the structure through relithiation, it not only reduces energy consumption and carbon emissions, but also yields lucrative high-value-added products (Figure 4B). Typically, direct regeneration technologies use hydrothermal lithiation to replenish lithium and incorporate a practical heat treatment to revive the crystal structure of the material. However, a challenge with this method is the influence of impurities on the regeneration process.<sup>[84,85]</sup>

Solid-state sintering is a strategy that uses lithium sources as raw materials, primarily for lithium replenishment. In this process, the lithium content in the cathode is precisely controlled, and a lithium source with a stoichiometric ratio (such as LiOH and  $\text{Li}_2\text{CO}_3$ ) is mixed with the cathode and regenerated at high temperatures. According to literature reports, various chemistries of LIBs, such as  $\text{LiCoO}_2$ ,  $\text{LiFePO}_4$ , and NMC, can be revitalized using this approach and show excellent electrochemical performance.<sup>[86–90]</sup> This approach is characterized by its simplicity, cost-effectiveness, and environmentally friendly nature; however, it necessitates precise measurement of the lithium content in the spent cathode. Given variations in restoration conditions, the devised process is applicable only to a specifically defined cathode material exhibiting similar degradation patterns. Moreover, the reduction in energy consumption and costs is facilitated by high-temperature sintering. The process of hydrothermal lithiation typically requires elevated pressure. To reduce the pressure required during regeneration, Shi et al. used a eutectic  $\text{Li}^{+}$  molten salt solution to lithiate spent NCM523, as shown in Figure 4E,F.<sup>[91]</sup> They innovatively combined low-temperature, low-pressure prelithiation and thermal annealing methods to successfully regenerate NCM523 with a discharge capacity of  $124.4 \text{ mAh g}^{-1}$  at 5C.

As shown in Figure 4C, Ji et al. employed a multifunctional organic lithium salt (3,4-dihydroxybenzonitrile dilithium) to directly regenerate degraded  $\text{LiFePO}_4$  cathodes from spent  $\text{LiFePO}_4$  batteries.<sup>[92]</sup> The degraded  $\text{LiFePO}_4$  particles were effectively coupled with the functional groups of the organic lithium salt, facilitating lithium filling vacancies and creating a reducing atmosphere to suppress the Fe phase. Concurrently, the thermal decomposition of the salt generated an amorphous conductive carbon layer that encapsulated the  $\text{LiFePO}_4$  particles, enhancing

the kinetics of lithium ion and electron transfer. The regenerated  $\text{LiFePO}_4$  cathode demonstrated outstanding cycling stability and rate performance, retaining 88% of its high capacity after 400 cycles at 5C. In another approach, Liu et al. introduced a simple, nondestructive, and triple-benefit solid-state sintering method for regenerating degraded  $\text{LiCoO}_2$  (D-LCO) cathodes, as depicted in Figure 4D.<sup>[93]</sup> This process enhanced the stability of D-LCO at high voltage, resulting in a cycled cathode that not only exhibited a high discharge capacity of  $188.2 \text{ mAh g}^{-1}$  at 0.2C but also maintained a capacity retention of 92.5% after 100 cycles (86.4% after 300 cycles) at a high cutoff voltage of 4.5 V, showcasing excellent rate performance.

Advantages of direct recycling methods: 1) simplified process, 2) the regenerated active materials can be directly used, and 3) reduction of secondary pollution. Disadvantages: the purity of the product cannot be guaranteed. The recycling process is susceptible to the influence of impurities. Currently, it is only applied on a laboratory scale and has not been scaled up for large-scale industrial applications.

## 5. Conclusion and Outlook

The metals found in LIBs, such as lithium, cobalt, nickel, among others, are finite resources. Recycling these metals plays a positive role in slowing down the overexploitation of mineral resources. Improper disposal of used LIBs may lead to soil and water pollution. Recycling can, therefore, reduce the negative impact of used batteries on the environment, in line with the concept of a circular economy. Conventional recycling methods involve multiple extraction and separation processes, which increase the overall recycling costs. In contrast, one-step regeneration methods streamline the process by avoiding the separation of various metal ions in the solution. After impurities are removed, the desired cathode material or high-value-added products are obtained by adding selected solvents to determine and adjust the filtrate concentration. This method significantly reduces energy consumption and carbon emissions. Although one-step regeneration methods have the potential for easy operation and high recycling value, addressing challenges, such as the impact of impurities on the regeneration process and the environmental effects of leaching agents, is crucial for large-scale implementation. In this regard, several recommendations are proposed.

**1) Internet integration:** Leveraging modern internet technology can optimize battery recycling by establishing a digital database for each battery batch from the production stage. Real-time monitoring of battery lifespan and automated reminders for replacement and recycling can ensure timely and efficient collection, reducing resource waste and energy input during regeneration. By fostering collaboration between logistics companies and battery manufacturers, a platform-based recycling network can be developed to streamline operations and link recycling points to enterprise management systems. Additionally, internet-driven solutions can support secondhand car transactions, battery replacements, and cascade utilization, further maximizing resource efficiency. The implementation of an internet-based

battery lifespan monitoring system not only enhances battery performance management but also reduces the need for manual classification during recycling, contributing to a more sustainable circular economy.

**2) Technological innovation:** Future efforts should focus on environmentally friendly and economically viable recycling technologies. While conventional multistep extraction processes are widely used, they consume large amounts of energy and generate secondary waste. In contrast, one-step regeneration offers a more efficient approach by directly recovering cathode materials while minimizing separation steps, reducing both energy consumption and emissions. However, key challenges, such as impurity control, reaction optimization, and material recovery efficiency, must be addressed. Advancing reaction mechanisms and refining process engineering will be crucial for large-scale implementation.

To further enhance sustainability, the development of green and efficient leaching and extraction agents should be prioritized. Replacing toxic and volatile organic solvents with biodegradable, nontoxic alternatives that decompose into harmless by-products can significantly reduce environmental risks. Process optimization should focus on improving efficiency while minimizing reagent consumption and energy use. Additionally, implementing closed-loop systems and automation can enhance safety by reducing operator exposure and ensuring controlled handling.

**3) Government involvement:** Robust policy frameworks are fundamental to accelerating LIB recycling at scale. Stricter environmental regulations, extended producer responsibility programs, and financial incentives such as subsidies or tax benefits can stimulate investment in advanced recycling technologies. Moreover, global collaboration will be essential for harmonizing recycling standards and managing cross-border material flows. Establishing a transparent and well-regulated market will help curb unauthorized recycling activities and ensure long-term industry growth.

In conclusion, LIB recycling holds profound significance for environmental and resource protection, offering new opportunities for economic development and advancing sustainable societal growth. Looking ahead, through global collaboration and technological innovation, the battery recycling industry is poised to become a key driving force for clean energy and green development.

## Acknowledgements

H.C. and L.-K.Z. contributed equally to this work. This work was supported by the National Natural Science Foundation of China (grant no. 52272194), LiaoNing Revitalization Talents Program (grant no. XLYC2007155), and the Fundamental Research Funds for the Central Universities (grant nos. N2025018 and N2025009).

## Conflict of Interest

The authors declare no conflict of interest.

**Keywords:** cathode materials • eco-friendly • lithium-ion batteries • one-step recovery and regeneration technology • recycling methods

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Manuscript received: December 21, 2024

Revised manuscript received: March 25, 2025

Version of record online: March 28, 2025