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Challenges and Opportunities of Recycling Cathode Materials from Spent Li-Ion Battery

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***Abstract*—With the rapid development of Electric Vehicles (EVs), the demand for Lithium-Ion Batteries (LIBs) has surged. However, the service life of LIBs is limited, and large-scale spent LIBs bring significant waste management and environmental challenges. Consequently, recycling spent LIBs has become an essential trend of resource recovery, environmental conservation, and social needs. The cathodes of LIBs contain valuable and resource-intensive materials like Lithium (Li), Cobalt (Co), Nickel (Ni), etc., which have substantial economic value and are a key focus in the recycling of LiB cathode materials. This paper provides an overview and assessment of conventional methods for recycling and reusing LIB cathode materials. The traditional LIBs recycling methods often involve extracting metal elements under high temperatures, and strong acid or alkali conditions, which might damage the environment easily, accompanied by high cost. Subsequently, the paper explores the advantages and disadvantages of direct recycling technology of cathode materials, compared with that of traditional recycling technology in process complexity, energy consumption, greenhouse gas emission, and cost considerations. Direct recycling technology emerges as the most favorable strategy for cathode material recycling, offering advantages in terms of cost, energy efficiency, and environmental impact. Finally, the strategy and challenges of direct cathode regeneration are summarized and discussed from the technical and environmental perspectives, and provide a new perspective for closed-loop recycling, to promote the industrialization and sustainable upgrading of cathode direct recycling.**

***Keywords*—spent lithium-ion batteries, cathode materials, recycling, direct cathode regeneration**

I. INTRODUCTION

LIBs have the advantages of high energy conversion efficiency, long cycle life, high energy density, and high power density and have been widely used in the field of EVs [1]. With the continuous growth of the EV market, an increasing number of LIBs are produced. Since LIBs contain Lithium (Li), Cobalt (Co), Nickel (Ni), Manganese (Mn), and other metals, as well as corrosive electrolytes and harmful components like inorganic and organic fluoride, if they are not properly disposed of, not only cause great space occupation pressure, but a huge amount of spent LIBs, in the long run, will also cause serious pollution to the atmosphere, soil, water and so on, meanwhile, causing great harm to the ecological environment and human health. It is urgent and important to carry out harmless treatment of spent LIBs to reduce environmental pollution.

Simultaneously, several crucial raw materials for LIBs are scarce and non-renewable. For example, Li, the main raw material of LIBs, is geographically concentrated, with 80% of the lithium ore concentrated in Australia and Chile, most other countries have to obtain it through imports, making LIB recycling crucial for mitigating lithium resource shortages. Co, another vital LIB component, about 70% of the world's Co mines are in the Democratic Republic of the Congo (DRC) [2]. Most of the DRC's cobalt supply comes from its industrial mining operations, which are plagued by ethical issues such as child labor and unsafe working practices. In addition, the extraction of Li and Co demands substantial energy and water, exacerbating resource consumption and environmental pressures. Ni in LIB cathode materials is also a valuable resource, often extracted from high-purity sulfide ores, which are mainly distributed in Russia, Canada, and South Africa. What's more, sulfur dioxide pollution during the mining process is also worrying [3]. Promoting LIB recycling is imperative to reduce excessive resource consumption, as well as realizing the reuse and sustainable development of resources.

Given this background, research and development of LIB recycling have made substantial progress over the past decade. With various methods, ranging from traditional pyrometallurgical and hydrometallurgical separation to the direct regeneration and utilization of LIBs, a systematic, green, and efficient LIB recycling system has gradually formed. Nevertheless, several challenges remain. For example, there still exist technical barriers that only better recycle the pre-materials of LIBs can be produced for the time being, uncertain dangers in the LIBs recycling process have not been grasped, and the legislative system of the LIB recycling industry requires further improvement.

This paper aims to delve into these issues, offering effective solutions. It will examine the evolution of battery recycling and treatment technology, as well as describe the characteristics, advantages, and drawbacks of traditional pyrometallurgy, hydrometallurgy, and direct regeneration methods. It will also compare and analyze these methods in terms of economic feasibility, environmental impact, operational efficiency, and complexity. The study will assess whether factors such as cost and carbon emissions are genuinely reduced through battery recycling. Finally, the paper will conclude with a summary and a forward-looking perspective, providing a valuable suggestion for advancing LIB recycling technology and maximizing the utilization of LIB resources.

II. TRADITIONAL RECYCLING TECHNOLOGY OF SPENT LITHIUM-ION BATTERIE

The traditional approach to recycling of LIBs primarily focuses on the extraction of lithium and various valuable metal elements, and the goal of process development is to enhance the extraction and leaching rate of metals. In both industry and academia, the traditional recycling process

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techniques that have been prominent include pyrometallurgy, hydrometallurgy, and physical and biological recycling. These methods are inadequate to a certain degree from the current perspective, but when the process of LIBs recycling develops, they are not only the representatives of the gradual development of battery recycling technology but also, they are the cornerstone of recycling technology upgrade research.

A. Pyrometallurgical Recycling

Pyrometallurgical recycling is a heat-based method that involves putting discharged LIBs to high-temperature calcination to extract valuable metal or non-metallic elements. This process entails placing spent LIBs into a melting furnace with a temperature higher than 100 ℃ for smelting. During smelting, organic matter such as the diaphragm, electrolyte, binder, and the anode’s graphite are incinerated and removed and removed. Metals with melting points lower than the reaction temperature are fused into alloys, while metals with low boiling points and their compounds are recycled through condensation, and other impurities are either directed to the slag box or transformed into gas [4].

Pyrometallurgical recycling has been industrialized in Umicore from Belgium [5]. The Val’Eas process involves introducing spent LIBs into three shaft furnaces operating at distinct temperature zones: the preheating zone (below 300 ℃), the plastic pyrolysis zone (around 700 ℃), and the smelting reduction zone (1200–1450 ℃). Through preheating discharge treatment and high-temperature roasting, the valuable metals within the battery are melted and reduced. Notably, Ni, Co, Copper (Cu), and iron (Fe) components are retrieved in the form of alloy products; While Li, Aluminum (Al), Silicon (Si), Calcium (Ca), and some Fe enter the slag box in the form of slag. Subsequently, Li needs to be further cooled and leached to form lithium salts in the pickling solution for recycling.

Pyrometallurgical recycling can be mixed with different types of spent LIBs, boasting significant processing capacity and a short recycling process. However, it does have certain drawbacks. Firstly, it is high energy consumption and high emissions. The high-temperature furnace calcination of metals and plastics generates a large amount of solid wastes, such as smelting slag and dust, as well as emissions like sulfur dioxide (SO_2), nitrogen oxides (NO and NO_2), and other waste gases which contribute to secondary atmosphere pollution [6]. Secondly, selective recycling of valuable metals can be challenging, particularly in the case of lithium, which necessitates further refinement; Thirdly, pyrometallurgical recycling primarily represents a destructive method, disorder of the original structure of the battery material, and leading to resource wastage and inefficiencies.

B. Hydrometallurgical Recycling Method

This method refers to the treatment of spent LIBs with chemical reagents such as acid, or ammonia immersion, and then extraction or separation of valuable metal elements in the solution by leaching or precipitation, to obtain the soluble metal salts or precipitate and other products. Hydrometallurgical recycling encompasses several stages, including pretreatment, leaching, purification separation, and material regeneration.

1) Acid leaching

Acid leaching can be carried out using either inorganic acids or organic acids. Inorganic acid leaching is a mature extraction technique and commonly used inorganic acids for leaching positive electrode waste materials including hydrochloric acid (HCl), sulfuric acid (H_2SO_4), or nitric acid (HNO_3), among others. HCl is a reducing agent, and no additional reducing agents are needed during leaching, while H_2SO_4 requires the addition of hydrogen peroxide (H_2O_2) for leaching.

Li *et al.* [7] employed HCl for leaching lithium cobalt oxide positive electrode materials. Under the conditions of 4 mol/L HCl, a leaching temperature of 80 ℃, and a leaching time of 2 hours, the leaching rates for Co and Li were 99% and 97%, respectively. $H_2SO_4 + H_2O_2$ system have been used to leach ternary positive electrode waste materials, achieving leaching rates of nearly 100% for Ni, Co, and Li, and a leaching rate of 94% for Mn [8].

Zhang *et al.* [9] summarized the effects of different factors, including acid concentration, temperature, time, solid-liquid ratio, and reducing agent dosage, on the leaching rate of valuable metal ions in spent LIB cathode materials. The conclusion is that the inorganic acid leaching of spent LIBs has the preponderance of easy control and a high metal leaching rate. However, strong acids such as H_2SO_4, HCl, or HNO_3 are highly corrosive, which has high requirements for equipment and produce substances such as SO_2 or chlorides (Cl2), which can lead to the secondary environment. The leaching rate of spent LIBs with organic acid can reach the same level as that with inorganic acid, but it is difficult to popularize and apply it industrially due to cost and other factors.

2) Ammonia leaching

Valuable metals like Li, Ni, Co, Cu, and ammonia have strong complexation abilities, forming stable metal complexes. In contrast, Fe, Al, and other metal ions have weaker complexation abilities when reacting with ammonia, and are difficult to dissolve into the solution by ammonia, to achieve selective leaching separation of metal elements.

Wang *et al.* [10] for the first time, demonstrated that ammonia leaching of spent LIBs exhibited good selectivity. After that, Wu *et al.* [11] studied the leaching behavior of Li, Ni, Co, Cu, and Al in the ammonia ($NH_3 \cdot H_2O$)-ammonium sulfite ($(NH_4)_2SO_4$)-ammonium bicarbonate (NH_4HCO_3) leaching system. They found that the changes in the concentration, leaching time, and temperature of (NH4) ₂SO₄ and NH_4HCO_3 were conducive to the leaching effect of Li, Ni, and Co. Under optimal conditions of 60 ℃, 1.5 mol/L $NH_3 \cdot H_2O$, 1 mol/L $(NH_4)_2SO_4$, 1 mol/L NH_4HCO_3, pulp density of 20 g/L and leaching time of 180 min, Ni and Cu leaching rates approached 100%, and Li and Co leaching rates reached 60.53% and 80.99%, respectively. The conclusion is that ammonia leaching exhibits high recovery efficiency. However, ammonia is volatile, as a result, making it operationally challenging and leading to the production of ammonia nitrogen wastewater, posing environmental risks. Therefore, how to realize the recycling of ammonia and avoid secondary pollution is the key issue to be considered in the industrialization of the ammonia leaching process.

Hydrometallurgy has the advantage of high metal recycling, but the utilization of strong acids, alkali, and other pollution, long process also being the shortages.

C. Physical Recycling Method

Contact electrocatalytic recycling technology is one of the typical physical recovery methods, which is based on the physical phenomenon of material contact electrification and the interdisciplinary integration of catalysis, for the spent LIB recycling.

Li *et al.* [12] proposed a method wherein silica acts as a catalyst, driven by mechanical energy, this could be used to induce superoxide free radicals, hydrogen peroxide, and other active substances by electron transfer generated by contact with water. This process effectively reduces high-value metals in electrode powder, facilitating the efficient leaching of Li, Ni, Mn, Co, and other metals. Experimental results show that under the condition of 90 ℃ and ultrasonic for 6 hours, the leaching rate of Lithium Cobalt Battery (LCB) reaches 100%, and the leaching rate of Co reaches 92.19%. In the case of a ternary lithium battery, under the condition of 70 ℃ and 6 hours, the leaching efficiency of Li, Ni, Mn, and Co are 94.56%, 96.62%, 96.54%, and 98.39%, respectively.

Due to the low cost of silica, when it acts as a dielectric powder catalyst, it is suitable for potential large-scale commercial applications. Moreover, the overall recycling process involves simple centrifugal separation, which means that the costs are further reduced. What’s more, contact electrocatalytic recycling technology can be classified as an environmentally friendly, cost-effective, and efficient approach to recycling spent LIBs.

D. Biological Recycling Technology

Biological recycling technology explores the use of microorganisms to catabolize battery materials and selectively leach specific elements. However, this approach is still in its infancy and lacks practical applications. Pollmann *et al.* [13] have used microorganisms to selectively digest metal oxides on the cathodes, reducing these oxides to metal nanoparticles. This emerging technology holds promise for LIB recycling and metal extraction, potentially complementing the established hydrometallurgical and pyrometallurgical processes currently used for metal extraction. Nevertheless, there have been limited studies in this area so far, and there are many opportunities for further research in this area.

III. DIRECT REGENERATION TECHNOLOGY OF SPENT LITHIUM-ION BATTERIES

A. Shortcomings of Traditional Recycling Technologies

Traditional recycling processes, particularly the commonly used pyrometallurgical and hydrometallurgical approaches, rely on dismantling the electrode material structure to extract valuable metal elements. Due to the stable structure of the electrode material, these methods necessitate extreme conditions, such as high temperature and strong acid or alkali, or ammonia treatment, to destroy the chemical bond in the electrode material. These recycling technologies also face a long chain of processes, high energy consumption, and large pollution, as well as the cost and emission of recycled additional reagents unable to be controlled, meanwhile, the application of recycled products has limitations and economic challenges.

B. Discussion and Advantage of the Emerging Direct Cathode Regeneration Technology

At present, academic research has identified two primary reasons for the degradation of LIBs:

(1) The lithium ions originally in the positive electrode cannot completely return to the original position during the battery charging and discharging cycle, and the thickened Solid Electrolyte Interface (SEI) is formed, resulting in the loss of lithium ions and the formation of lithium defects in the positive crystal, resulting in the reduction of battery capacity [14].

(2) Long-term contact with the electrolyte causes phase transition on the cathode material surface, reducing lithium ion conductivity and increasing polarization, resulting in decreased battery capacity [15]. Aiming at the causes of LIB function degradation, direct regeneration technology that directly supplements Li to restore LIB performance came into being.

1) Current status of the direct cathode regeneration technology

Direct regeneration technology is a straightforward recycling technology that restores the performance of LIBs by replenishing lithium to repair the active material of the battery electrode. It mainly uses lithium precursors (e.g., LiOH, Li_2CO_3, LiCl, LiBr) as Li supplements, calcines the recrystallized cathode at high temperature to generate regenerated cathode powder, and directly mixes the obtained regenerated cathode materials (lithium cobalt, lithium manganate, lithium iron phosphate, and nickel-cobalt-manganese) with LiOH or Li_2CO_3 at an appropriate molar ratio to restore the lost Li and the crystal structure, thus returning the electrochemical properties to their original state. The direct regeneration method abandons the mind of destroying particles in pyrometallurgy and hydrometallurgy, which separate the cathode materials from the spent batteries by physical methods and then replenish lithium to the attenuated cathode materials. The treated regenerated cathode materials exhibit high capacity, and excellent cycling performance, and can be directly used in new batteries. Direct regeneration methods encompass thermal repair and regeneration and hydro-chemical/electrochemical re-lithiation.

Thermal repair regeneration: Thermal repair regeneration is used to repair lithium-deficient cathode materials by supplementing the lithium source and heating. The underlying mechanism diffusion of lithium ions into the sites with lithium defects in the spent LIB cathode material to achieve regeneration of the spent active material. This method yields regenerated materials with exceptional crystallinity and desired stoichiometry. Thermal repair regeneration includes two main processes:

(1) Solid-state sintering process: Several solid-state sintering processes have been explored for recycling waste cathode materials like $LixCoO_2$, LixNi1/3Co1/3Mn1/3O2, and $LixFePO_4$. For instance, Nie *et al.* [16] utilized a solid-state synthesis process to regenerate $LixCoO_2$ using waste cathode materials.

(2) Eutectic melting process: This technique involves the

preparation of low eutectic molten salt solution to lower the melting point and reduce greenhouse gas emissions, Li+ ions in the eutectic molten salt can easily compensate for the lithium defect vacancies in the spent LIB, allowing re-lithiation of cathode without the need for additional pressure. It has been shown that the addition of Ni compounds to the Reciprocal Ternary Molten Salt (RTMS) system can facilitate re-lithiation and Ni enrichment processes for waste NCM111 materials [17]. This process directly transforms waste NCM111 to Ni-rich NCM (Nr-NCMS). Eventually, the lithium defective vacancies of the waste NCM111 are repaired and the obtained Nr-NCMS material exhibits an α-$NaFeO_2$ type layered structure and excellent properties similar to those of the fresh NCM622. The RTMS system can significantly reduce the eutectic melting temperature down to 300 ℃, enhancing the efficiency of flux processes.

Hydro-chemical re-lithiation: In addition to the thermal repair of regenerated cathode materials, the direct recycling of spent LIB cathodes can also be achieved in an aqueous solvent environment. The re-lithiation process in a solvent environment requires less energy compared to thermal repair regeneration, contributing to a reduction in carbon emissions. This includes hydrothermal, ionothermal, and electrochemical re-lithiation.

(1) Hydrothermal re-lithiation: Hydrothermal reactions in lithium-containing solutions can be successfully used to generate particles with the desired stoichiometry and high crystallinity. For instance, Wang *et al.* [18] added recycled lithium manganate material to a polytetrafluoroethylene-lined autoclave filled with LiOH solution and heated it at 145–185 ℃, demonstrating that Li loss and lattice disorder can be fully recovered without an annealing process.

(2) Ionothermal re-lithiation: Ionic thermal re-lithiation can be viewed as an alternative to direct regeneration of cathode active material in a mild environment. Ionic liquids (ILs) are used to prepare solid-state materials with minimal morphological changes, serving as a new reaction and fluxing medium. Wang *et al.* [19] designed a process to relitigate ternary lithium battery by ionothermal synthesis at 150–250 ℃ using recyclable ionic liquids as solvents and lithium halides as lithium salts, which exhibited excellent electrochemical performance. Ionothermal repair offers numerous advantages, such as negligible vapor pressure, non-flammability, high thermal stability, and excellent flexibility in synthesis performance.

(3) Electrochemical re-lithiation: Electrochemical re-lithiation regeneration technology has gained increasing attention. The mechanism is that the electrons output from the electrical energy supply device to the negatively charged cathode through the conductive medium, and lithium ions are eventually successfully replenished to the lithium-deficient sites of the spent $LixCoO_2$ in the electrochemical drive when the energy in the electrochemical system is greater than the activation energy-limit. It’s worth noting that many Li ions that do not enter the lithium defect sites of the spent $LixCoO_2$ are likely to be inserted into the interlayer gaps of the $LixCoO_2$ layered structure. The formation of hydrogen bonds leads to the generation of $LiCoO_2$ particles during electrochemical re-lithiation regeneration. The charging capacity of the regenerated material is 136 mAh g-1, which is almost comparable to the charging capacity of commercial $LiCoO_2$ (140 mAh g-1) [20]. However, the electrochemical reaction is prone to side reactions and requires matching electrochemical devices as well as preparation and separation of electrodes, making it unsuitable for large-scale industrial regeneration. Generally, circulating water thermal lithiation can be used to regenerate cathode materials with fewer phase changes and superior electrochemical performance.

2) *Discussion of the direct cathode regeneration technology*

From the above analysis and discussion of emerging direct cathode material regeneration technologies in recent years, it is evident that the high-temperature energy input required for the solid-state sintering process plays a pivotal role in achieving crystal structure regeneration and re-lithiation, which implies a higher energy consumption. In addition, the precise control of the stoichiometric ratio of the regenerated cathode material is difficult to achieve, resulting in poor compositional homogeneity of the solid-phase interface of the regenerated material, and thus a high failure rate of the re-lithiation effect of the raw material. Compared with the solid-state sintering process, although thermal eutectic re-lithiation has made a breakthrough in the lithiation temperature/time/solid-phase surface homogenization, a great challenge in the lithium supply metrology and the strict control of the lithiation time still exists, which leads to the need to improve the applicability of the cathode materials with different health grades. In summary, the restoration of lithium vacancies in the cathode, whether through thermal repair regeneration or hydrochemical/electrochemical lithiation, is rooted in addressing the cathode failure mechanism of LIBs.

Despite persistent technical challenges in these methods, such as the unmet goals related to the initial discharge capacity and cycling performance of regenerated materials when compared to new materials, the direct regeneration approach represents a significant advancement over traditional recycling methods (hydrometallurgy and pyrometallurgy) in terms of preserving the original material structure. Therefore, we further discuss in depth the advantages and disadvantages of conventional recycling technologies and direct cathode material regeneration technologies in terms of process complexity, material flow, recycling quality, energy consumption, greenhouse gas emissions, and cost in the following sections.

C. *Comparison of Recycling Methods in Process Complexity, Material Flow, and Recycling Efficiency*

As shown in Fig. 1, Wang *et al.* [21] compared the material flow among the recycling processes of direct recycling, traditional pyrometallurgy, and hydrometallurgy methods. Figs. 1a, 1c, and 1e demonstrate that the direct recycling method significantly reduces process time, minimizes the use of acid and alkali reagents during the restoration process, substantially reduces harmful emissions, and outperforms traditional pyrometallurgy and hydrometallurgy methods. Direct recycling restores obsolete waste materials to a good structure, and the performance of the restored materials is as good as or even better than new batteries. This method is simple and economically beneficial, as depicted in Fig. 1f.

Direct recycling emerges as a highly promising recycling method with substantial practical application potential.

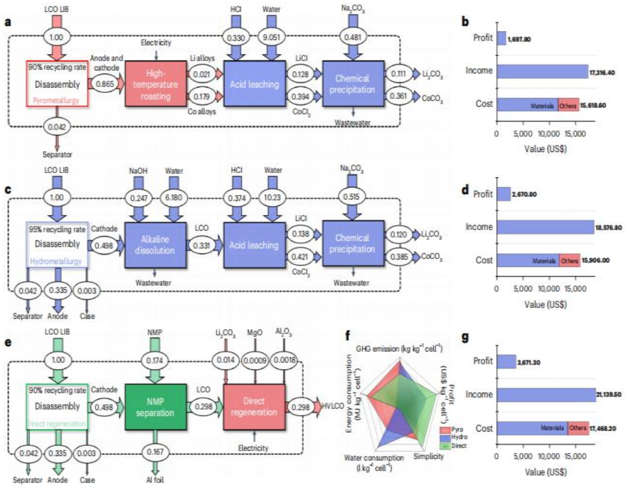

Fig. 1. (a) Material flow analysis of pyrometallurgy recycling process. (b) Economic analysis of pyrometallurgy recycling process. (c) Material flow analysis of hydrometallurgy recycling process. (d) Economic analysis of hydrometallurgy recycling process. (e) Material flow analysis of recycling methods. (f) Comprehensive comparison of direct recycling, pyrometallurgy recycling, and hydrometallurgy recycling. (g) Economic analysis of direct recycling. Numbers in ellipses in a, c, and e represent the mass of each material. GHG, a greenhouse gas; HVLCO, high-voltage LCO; NMP, 1-methyl-2-pyrrolidinone [21].

In Fig. 2, a comparative analysis was conducted, assessing the technical readiness, process complexity, quality and quantity of recycled metals, waste generation, and the direct usability and extraction rate of metal/non-metal elements in three different LIB recycling methods: pyrometallurgy, hydrometallurgy, and direct recycling. Notably, direct recycling excels in several indicators.

Comparison of different LiB recycling methods

Best ••••• •••• ••• •• • Worst

	Technology readiness	Complexity	Quality of recovered material	Quantity of recovered material	Waste generation	Energy usage	Capital cost	Production cost
Pyrometallurgy	•••••	•••••	•	•••	••	•	•	•••••
Hydrometallurgy	••••	•••	•••	••••	•••	•••	•••	•••
Direct recycling	••	•	••	•••••	••••	•••	•••	•

	Presorting of batteries required	Cathode morphology preserved	Material suitable for direct re-use	Cobalt recovered	Nickel recovered	Copper recovered	Manganese recovered	Aluminium recovered	Lithium recovered
Pyrometallurgy	•••••	No	No	•••••	•••••	•••••	•••	No	•
Hydrometallurgy	••••	No	No	•••••	•••••	••••	•••	•••••	•••
Direct recycling	•	•••••	••••	•••••	•••••	•••••	•••••	•••••	•••••

Fig. 2. Comparison of different LIB recycling methods [22].

D. Comparison of Recycling Methods in Energy Consumption, Greenhouse Gas Emissions, and Costs

EverBatt conducted a simulation of a comparative model examining the lifecycle energy consumption, greenhouse gas emissions, and costs for the recycling processes of spent LFP cathodes using pyrometallurgy, hydrometallurgy, and direct recycling techniques. The model was developed by Argonne National Laboratory [20], as shown in Fig. 3.

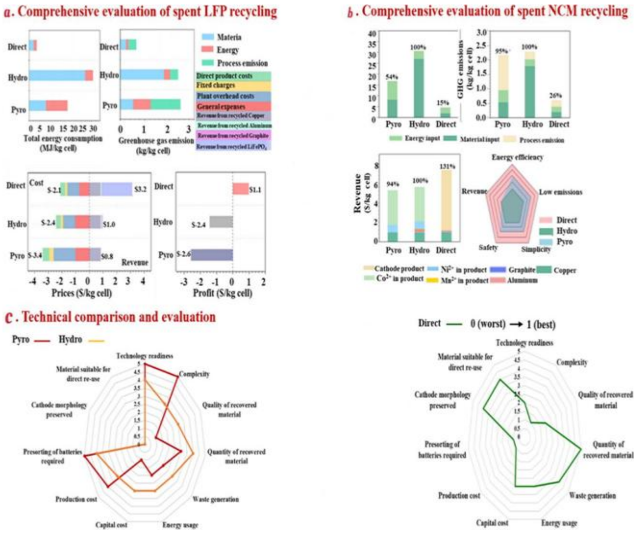

Fig. 3. (a) Schematic illustration of the comprehensive evaluation of spent LFP recycling: Total energy consumption, greenhouse gas emission, prices, and profit for recovering 1 kg of waste LIBs by pyrometallurgy, hydrometallurgy, and direct regeneration recycling; (b) Comprehensive evaluation of spent NCM recycling: Energy consumption, greenhouse gas emissions, potential revenue (economic benefit) from outputs produced, Comparison of different LIBs recycling methods. (c) Technical comparison and evaluation of pyrometallurgy, hydrometallurgy, and direct regeneration process, according to the aspects of material suitable for direct re-use, complexity, cathode morphology preserved, presenting of batteries required, production cost, capital cost, quality, quantity of recovery material, waste generation, and energy usage. Pyro, Hydro, and Direct represent pyrometallurgy, hydrometallurgy, and direct regeneration process, respectively [20].

For recycling 1 kg of spent LFP cathodes (Fig. 3a), the total lifecycle energy consumption for pyrometallurgy and hydrometallurgy is 18.4 MJ and 30.6 MJ, respectively. In contrast, direct recycling consumes only 3.5 MJ, significantly lower than pyrometallurgy and hydrometallurgy. Notably, a substantial portion of the energy consumption for pyrometallurgy comes from high-temperature smelting, while 87.8% of hydrometallurgy's energy consumption arises from upstream chemical reagent manufacturing. Greenhouse gas emissions for direct recycling account for 26.6% and 27.7% of those for pyrometallurgy and hydrometallurgy, respectively. In the case of direct recycling, the cost of chemical lithium salts is the main expense and is significantly lower than that of pyrometallurgy or hydrometallurgy routes.

In the direct recycling process, the sole raw material used is LiOH, the amount of which depends on the loss of lithium in the waste NCM cathode materials. The reduction in chemical reagent consumption and process requirements is driving the development of direct recycling. The energy consumption for recycling 1 kg of waste NCM is reduced to 4.5 MJ, constituting only 15% of hydrometallurgy's energy consumption. Direct recycling of 1 kg of NCM only releases 0.6 kg of greenhouse gases, which is significantly lower than pyrometallurgy (2.16 kg) and hydrometallurgy (2.27 kg). In addition to minimal energy consumption and greenhouse gas emissions, the direct recycling product can be used directly to manufacture new LIBs, creating greater economic value compared to pyrometallurgy and hydrometallurgy. It is estimated that recycled cathodes, represented by pale yellow bars, will be converted to $5.80 and yield $6.90 in total economic benefits. However, in the case of pyrometallurgy and hydrometallurgy, cathodes degrade into simple compounds. While Ni and Co precursors hold high value, the overall economic benefit generated by pyrometallurgy and hydrometallurgy is estimated to be only $5.20/$5.60. If the final output products of pyrometallurgy and hydrometallurgy are the same as those obtained from direct recycling, the energy consumption and greenhouse gas emissions generated will undoubtedly be higher than this value.

From a technical perspective (Fig. 3c), pyrometallurgy does not exhibit significant advantages. It demonstrates higher indicators in terms of technical maturity, required battery pre-sorting, and complexity, but exhibits lower indicators regarding the quantity and quality of recycled materials. As a result, this method has higher energy and environmental costs. Hydrometallurgy performs better than pyrometallurgy but still has room for improvement. Direct lithium recycling technology surpasses the two traditional recycling methods in terms of technology, cost, and products. These indicators suggest that direct lithium recycling technology offers a simpler process, lower consumption, and higher productivity, which means it will consume less energy and have lower environmental costs, ultimately maximizing economic benefits.

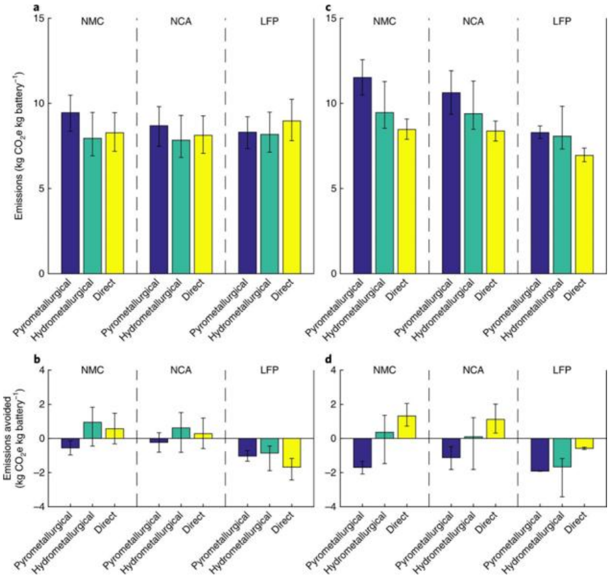

Fig. 4. Diagram (a) illustrates the CO_2 equivalent emissions of cylindrical batteries made and recycled through pyrometallurgy, hydrometallurgy, and direct cathode recycling methods for the average U.S. electricity grid. Diagram (b) shows the median values along with 95% confidence intervals for greenhouse gas emission reductions using each recycling process. Diagrams (c) and (d) show the results for pouch cells [23].

As shown in Fig. 4, for LFP batteries, all considered recycling methods and battery formats result in a net increase in CO_2 emissions when considering the incineration of materials of no value, including transition metals, lithium, or cathode materials, or as waste (battery hardware and current collectors). In comparison to Ni, Co, and Mn precursor cathodes, Fe precursor materials used in LFP cathodes have higher energy efficiency in mining and lower greenhouse gas emissions per kilogram and kilowatt-hour. This results in smaller greenhouse gas offsets from recycled materials, which are insufficient to counterbalance the energy and greenhouse gas emissions associated with the recycling processes. For NMC and NCA batteries, both hydrometallurgy and direct cathode recycling result in reduced greenhouse gas emissions. However, these reductions are statistically significant only for pouch cells recycled through the direct cathode recycling process. Pyrometallurgy recycling causes a net increase in greenhouse gas emissions and energy consumption, particularly when considering the results per kilowatt-hour of battery recycling. Hydrometallurgy and direct recycling have the potential to diminish greenhouse gas emissions and energy consumption for NMC and NCA batteries.

To make direct cathode recycling have a smaller greenhouse gas impact than pyrometallurgy or hydrometallurgy recycling, the cathode recovery rate from direct recycling depends on the battery format and chemistry. The breakeven cathode recovery rate for direct cathode recycling exceeds the performance of hydrometallurgical chemistry and pyrometallurgy recycling for cylindrical cells. These considerations also apply to pouch cells. Since hydrometallurgy recycling processes can achieve moderate reductions in CO_2 emissions, high cathode recovery rates are essential for direct cathode recycling to achieve significant CO_2 offsets (all indicators >40%). However, because pyrometallurgy recycling leads to a net increase in greenhouse gas emissions, even a lower cathode recovery rate in the direct cathode recycling process still results in lower greenhouse gas emissions compared to pyrometallurgy recycling.

IV. CONCLUSION

Looking at the evolution of LIB recycling, continuous progress in academic research and industrial applications drives the development of recycling technologies. This article conducts a comprehensive analysis, considering technical, environmental, and economic perspectives of the three primary recycling methods, and draws the following conclusions:

Direct cathode material regeneration, as an advanced recycling technology, introduces innovative approaches to address lithium deficiency and structural defects in LIBs. It offers three significant advantages: (1) Substantial reduction in process time. (2) Reduced use of chemical reagents during LIB recycling, leading to a significant decrease in greenhouse gas emissions and secondary pollutant emissions, with a notable impact on reducing environmental pollution. (3) The directly regenerated materials can be used to manufacture new LIBs, saving on resource consumption and providing significant economic benefits.

Direct regeneration technology is superior to pyrometallurgy and hydrometallurgy processes and holds substantial promise as a practical and applicable recycling method.

However, in real-world applications, direct recycling technology faces certain difficulties and challenges. such as: (1) The direct regeneration process must be customized for specific positive electrode materials. Even for the same type of positive electrode material, different health conditions require different process parameters and lithium supply reagents, including the selection of single or mixed types and quantity optimization. (2) The efficiency of the direct regeneration process is related to the health status of the LIBs, and a low state of charge does not favor direct recycling. (3) The direct regeneration process is difficult to adapt to source materials of unknown or poor quality. If the product quality is compromised, companies may be hesitant to use recycled materials again.

Addressing these challenges means that corresponding industrial integration and testing technologies must be developed. This is essential for upgrading direct regeneration technology to further reduce costs and energy consumption.

Given the existing bottlenecks and challenges in environmental and technical aspects of LIB recycling processes, exploring more streamlined, efficient, and environmentally friendly diversified recycling technologies has become the basis for the healthy and sustainable development of the spent LIB recycling industry and related upstream and downstream industries.

CONFLICT OF INTEREST

The author claims that no conflict of interest exists.

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