

Challenges and Perspectives for Direct Recycling of Electrode Scraps and End-of-Life Lithium-ion Batteries

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The growing demand and production of lithium-ion batteries (LIBs) have led to a critical concern regarding their resources and end-of-life management. Consequently, LIB recycling has emerged as a prominent topic in academia and in industries, driven by new worldwide governmental regulations and the increasing gap between the supply and demand of critical and strategic raw materials. Widely considered as a more sustainable and cheaper recycling method compared to pyrometallurgy and hydrometallurgy, direct recycling currently grabs the spotlight. This perspective provides insights and outlooks on the chemical and technological challenges of the innovative direct

recycling approach for LIBs, addressing both the production scraps and batteries at their end-of-life (EOL). Technological advancements, changes in battery chemistry, along with the LIB market dynamics and collaborations between battery makers and recyclers, are key drivers of LIB waste recycling. While production scraps lend themselves well to direct recycling, EOL batteries encounter challenges in adopting this novel recycling technology. Besides, the need to assess novel direct recycling processes using Life Cycle Assessment (LCA) is also important for identifying eco-design strategies and optimizing the processes, leading to a more sustainable energy storage system.

Introduction

Energy storage has emerged as a cornerstone of modern society's pursuit of a sustainable future. Lithium-ion batteries (LIBs) transformed the paradigm of powering our devices, from smartphones to electric vehicles (EVs), and have played a pivotal role in the transition to renewable energy sources. However, as our reliance on LIBs continues to grow, a pressing challenge has emerged: the responsible and efficient management of the exponentially increasing demand for raw materials and the wastes generated during the development and utilization of this technology.^[1]

The disposal and recycling of LIBs have become an urgent concern as their widespread use raises questions about resource conservation, environmental impacts, and energy security. New governmental regulations, such as the (EU) 2023/1542 of the European Union, set constraints on companies to identify, collect, and recycle spent batteries. They also mandate the reincorporation of a certain amount of strategic elements such as Cobalt (Co), Lithium (Li) or Nickel (Ni), into new batteries to further circularity.^[2] Similar regulations are practiced by other countries such as the USA and China encompassing economic, social, technological, and environmental aspects.^[3,4]

Traditional recycling methods namely pyro- and hydrometallurgy often involve expensive and resource-intensive processes aimed at essentially recovering valuable transition metals, leading to limited overall recycling efficiency.^[5] These methods, although rather efficient, are only profitable when a high quantity of elements such as Co or Ni are present in the urban mining; whose value is also dependent on their market price which is rather unpredictable, making its profitability, chemistry and market dependent. To increase recycling efficiency and minimize the environmental and economic footprints of the LIB recycling, direct recycling has emerged as an innovative and a more sustainable approach.^[5] Unlike traditional recycling methods that destroy LIBs to recover valuable elements, direct recycling aims at the recovery, regeneration, and reuse of battery components (active materials, current collectors, electrolyte, polymer binders, etc.) directly, without breaking down the chemical structure, or in other words preserving the energy in the material.

In recent years, numerous innovative methodologies were developed to extract the electrolyte, or to delaminate, recover and regenerate both negative and positive active materials, as illustrated in some recent review papers providing a current

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state-of-the-art of the lab-scale research.^[5–7] This perspective paper does not focus on the diverse approaches for recycling/regenerating the active materials constituting LIBs, nor addressing the potential of a second life. It has for objective to shed light on all the challenges that face direct recycling whether for electrode production scraps or end-of-life (EOL) batteries, and to propose some research directions to pursue (Figure 1).

EOL Batteries vs. Electrode Scraps

First, it is important to describe the characteristics of the different products that have to be recycled. EOL LIBs and production scraps represent distinct stages in the life cycle of batteries, each with its unique characteristics. EOL batteries refer to batteries that have reached the end of their useful operational lifespan and are no longer capable of providing reliable or efficient electrical energy storage. Typically, LIBs are considered at their EOL once their capacity retention drops to 70–80% of the initial capacity, which corresponds to a lifespan of 10–20 years for an EV battery.^[8,9] These batteries have experienced wear and degradation during usage, posing significant challenges in recycling due to the presence of deteriorated materials. Moreover, valuable components for recycling (e.g. positive and negative electrode materials, current collectors, etc.) are incorporated in cells assembled into battery packs, and thus, are not easily accessible. Additionally, proprietary knowledge regarding the content of these packs is often unavailable, for instance some companies mix cathode active materials for specific applications.^[10,11] These factors alone require various steps and pretreatments to get access to the degraded electrodes.

On the other hand, electrode scraps are generated during the manufacturing stage of batteries. These scraps can include imperfect or surplus components, excess materials, or defective units produced during the assembly. Unlike EOL, production scraps are relatively less complex, safer to recycle and easy to process as they have not undergone the wear and tear associated with actual usage. Besides, electrode production scraps can be directly collected at different stages of the



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production; hence its content/chemistry is known to cell producers and has no requirement of dismantling cells/packs or performing other complex manual operations. Therefore, direct recycling appears perfectly adapted for this type of waste. Direct recycling methods can efficiently recover valuable materials from electrode scraps, such as the active materials (LiFePO_4 (LFP), $\text{LiNi}_{1-x-y}\text{Mn}_x\text{Co}_y\text{O}_2$ (NMC), $\text{LiNi}_{1-x-y}\text{Co}_x\text{Al}_y\text{O}_2$ (NCA), graphite, graphite-silicon, etc.), the current collectors (Al and Cu) and other components, and reintegrate them into the manufacturing process. The controlled and consistent nature of production scrap materials allows a straightforward separation and processing techniques, contributing to the overall efficiency and effectiveness of direct recycling practices in the context of battery manufacturing.

Direct Recycling of Electrode Production Scraps

Recent studies have revealed that the amount of electrode production scraps can vary from 5 wt.% to 30 wt.% of the total production depending on the maturity and scale of factories, whether startups or gigafactories.^[5] Considering the overall production required for urban mobility electrification (350 GWh in 2024 and 1700 GWh in 2030 according to Bloomberg New Energy Finance), this represents an enormous quantity of electrode scraps that needs urgent recycling. Indeed, electrode scraps are generated at the production stage and thus, are readily available for recycling and reincorporation into the production line, whereas the influx of EOL batteries will arrive in the next decade, when the batteries will be degraded. To highlight this point, a report by Umicore suggests that compared to EOL batteries, production scraps will be the primary source of supply for the production of new LIBs, at least up to 2030.^[12]

Direct recycling of the electrode production scraps does not require discharging or other pretreatment steps, it immediately proceeds to separate the active material together with the electronic conductor additive from the current collector, either by removing the binder or by altering its cohesive/adhesive properties (Figure 1). The challenge of this step, both for cathode and anode, lies in perfectly delaminating the electrode to recover a mixture of carbon black (CB) and active material or a pure active material without altering its structure, properties and energy storage performance. It is important to note that a simple room-temperature stirring of the electrode scraps in the same solvent used for slurry preparation is usually inefficient to achieve a complete delamination. Therefore, various approaches have been investigated to achieve electrode delamination. For instance, thermal processes decompose the binder,^[13,14] whereas mechanical processes shred the electrode to detach the composite made of active material, CB, and binder from the current collector.^[15,16] However, in the former case, the binder may potentially react with the active material (and/or the current collector) during its decomposition,^[17] whereas in the latter approach, the binder remains present, covering the active material particles, and therefore, altering the quality of the recovered active material.^[18] Additionally, a

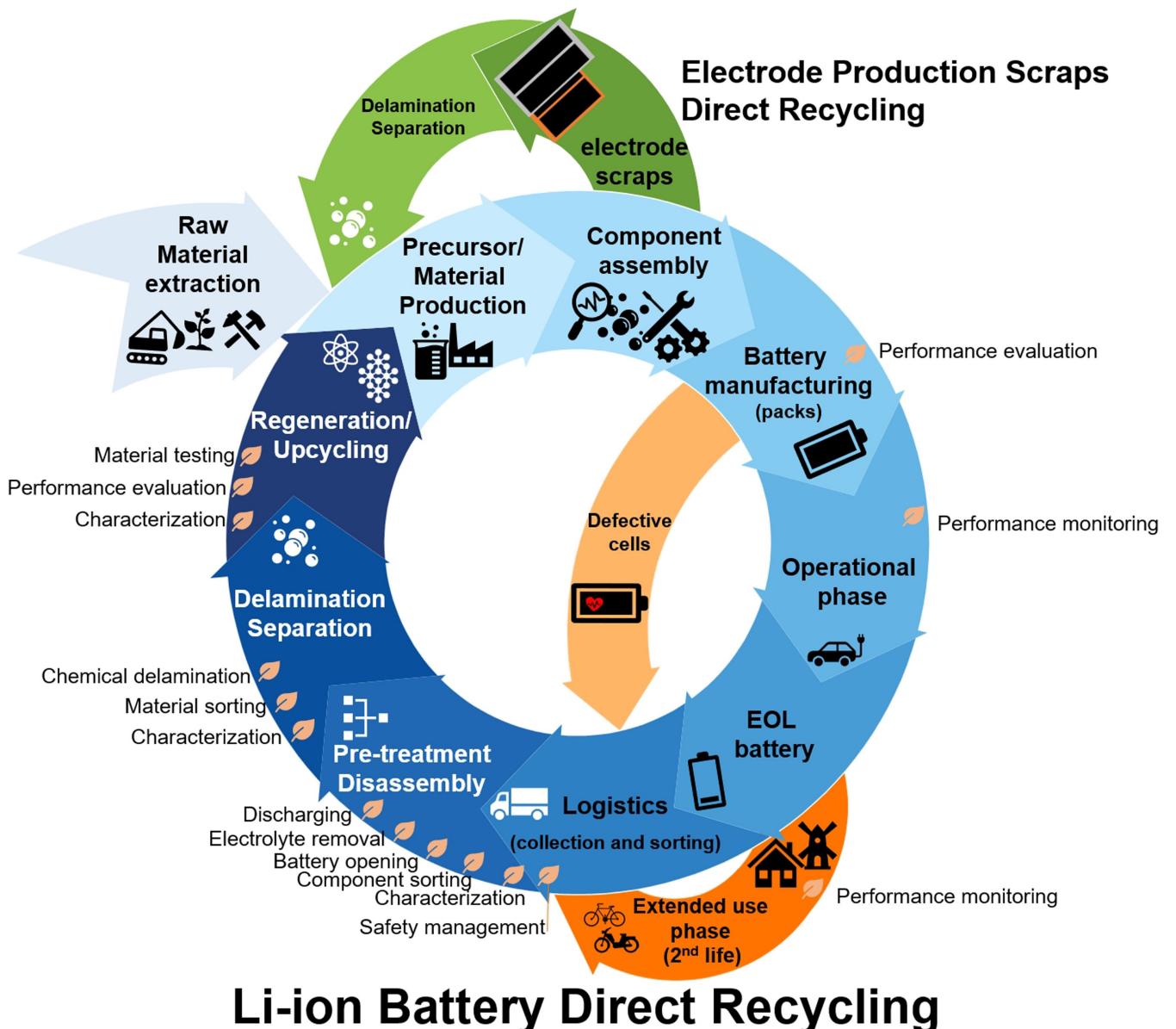


Figure 1. Lithium-ion battery and electrode scrap life cycle in the strategy of direct recycling.

high degree of comminution generates fine particles of copper (Cu) and/or aluminum (Al), requiring further separation steps from the electrode materials.^[18,19] Among the physico-chemical, mechanical or thermal processes investigated for this purpose, the physico-chemical approach, which involves dissolving the binder, appears to be the most promising.^[20–23] This method typically requires a simple and low-temperature process and low energy consumption, can be adapted to different cathode and anode chemistries and is the only one that allows to recover the binder. The selection of the solvent must conform to binder's properties and a green solvent that is inert to the active material should be favored.^[24] For instance, triethyl phosphate (TEP),^[21] ethylene glycol (EG),^[25] cyrene^[26] or dimethyl isosorbide (DMI),^[27] etc. were employed to dissolve PVDF (polyvinylidene difluoride) binder used in positive electrodes whereas water can be used to delaminate the negative

electrode.^[21,28] It must be noted that the industrial casting manufacturing processes are more optimized than lab-scale ones, leading to a more challenging delamination. Therefore, it is necessary, to the extent possible, to validate the experimental conditions using industrial electrode production scraps.

By this physico-chemical approach, the active material can usually be recovered together with the electronic conductor additive, typically CB. The difference in physicochemical surface properties between the active material and the CB enables their separation by flotation without destroying one of them. However, in practice, achieving complete material separation is challenging and time consuming, and it appears useless as both components are necessary for the slurry formulation. Additionally, contrary to mechanical approach, physico-chemical delamination often permits to recover a clean current collector, ready for downstream processing.

Although the comprehensive study of chemical composition and structure of the recycled active material is crucial, the investigation of its surface chemistry and microstructure should not be overlooked. For instance, traces of solvent or binder on the material's surface could degrade its performance. Partial exfoliation of graphite during the physico-chemical delamination process could also induce drastic effects on the energy storage performance of the recovered material. Moreover, certain aspects, often dependent on active material, are essential to assess the limitations of such direct recycling approach and to potentially further optimize the entire recycling process. For instance, when dealing with layered positive electrode materials, it is necessary to understand the effect of electrode processing on the material, and more specifically, on its microstructure (impact of calendering). Particle cracking during the calendering step would irreversibly alter the material, preventing the recovery of materials with the same microstructure as the pristine one. Therefore, to restore the microstructure, additional treatments could be performed, or adjustments of electrode formulation and slurry rheology might be necessary at an industrial scale to reach optimized energy storage performance. Ultimately, the success of direct recycling is evaluated by the quality and the energy storage performance of the recovered active material. In this frame, electrochemical impedance spectroscopy to probe materials resistivity, along with long-term cycling stability and evaluation of capacity retention at high current densities, are of high importance in better evaluating the efficiency of the recycling process. In addition, it is rather rare to see a new device that is made of 100% recycled material. Therefore, studies on testing the energy storage performance of mixtures containing recycled and pristine active materials should be further investigated.^[24] Several studies have successfully recovered various chemistries of electrode materials with performance similar to pristine ones, highlighting the promise and relevance of direct recycling for electrodes scraps.^[25] In parallel, start-ups focusing on direct recycling of electrode scraps see daylight, confirming the large-scale interest of such a recycling approach.

Direct Recycling of EOL Batteries

The direct recycling of EOL batteries already poses substantial challenges beyond the scientific domain. A complete direct recycling involves multiple stages, including collection, sorting, discharging and dismantling the batteries, opening the cells, extracting the electrolyte, delaminating the electrode materials from the current collectors, and ultimately regenerating the degraded electrode materials (Figure 1). Moreover, several steps of this full procedure introduce safety concerns for operators and machinery, such as risk of explosion during battery storage and transport (collection step) or dismantling step, and exposure to HF during battery opening.

The initial stages of the process present logistical and technological challenges, which can be partially addressed through the establishment of industrial standards for LIBs, the development of efficient human-machine hybrid workstations

or eco-conception approaches.^[6] On the other hand, challenges in electrolyte recovery and electrode material separation and regeneration require addressing chemical complexities. While only a few works have been performed on the electrolyte recycling, the majority of them employed sub- or supercritical CO₂ processes to extract and recover it.^[30-32] These studies performed either on separator soaked with electrolyte or on actual cells, proposed ways to valorize the electrolyte and showed the possibility for its recovery. However, none of them reported the re-use of recovered electrolyte in a new cell, which would be the ultimate goal to validate the process.

It is widely accepted that the electrolyte undergoes degradation during battery operation (Solid Electrolyte Interface (SEI) formation, LiPF₆ hydrolysis, etc.). Thus, its direct re-use in new cells is improbable. Moreover, regenerating the electrolyte is likely to pose significant challenges due to the volatility of solvents; it contains ethylene carbonate, dimethyl carbonate, etc. and the presence of various additives that irreversibly evolve during cycling. These phenomena may limit the possibility of electrolyte recycling and re-use. Therefore, we propose viewing electrolyte extraction as a pretreatment step to enhance safety and minimize contamination on recovered electrodes. In this regard, a comprehensive examination of the state of other battery cell components before and after electrolyte extraction is essential to get the full picture of the process and evaluate its efficiency.

The direct recycling of EOL electrodes, particularly positive electrodes, has garnered extensive attention in recent years due to the high cost of cathode materials compared to other components.^[33] Primarily, similar physico-chemical, thermal and mechanical delamination approaches as those used for electrode scraps are usually employed to separate the active material from metallic foils. It is important to mention that Bai et al. conducted chemical delamination to electrode scraps and EOL positive electrodes and observed a difficulty in translating the same process to degraded material.^[21] It was identified that the cathode electrolyte interface layer greatly decelerates the delamination. To avoid this initial delamination step, innovative direct recycling methods focused on the entire electrode are emerging, such as the study of Ouaneche et al. which successfully regenerated degraded LFP still attached on the aluminum foil.^[34]

One of the major challenges associated with EOL positive electrodes lies in regenerating the active material, especially when its degradation state is unknown. Various methods such as hydrothermal, ionothermal, redox mediation, electrochemical and solid-state regenerations have been explored for this purpose.^[6,7,35,36] However, much of the current research focuses on ideal and controlled scenarios where well-characterized degraded material merely necessitated a topochemical relithiation, followed by a post-annealing to eliminate potential residues and restore the microstructure. In real-word scenarios, inhomogeneities in terms of degradation can already exist within a single electrode film and are highly probable among different cells within the same EV battery pack. For instance, post-mortem analyses on an EOL 18650 cell revealed differences in chemical composition between the beginning, middle

and end of the cathode roll.^[37] Furthermore, it is unrealistic to assume that the degraded electrode material for each cell would be finely characterized at an industrial scale (composition, coating, structure, microstructure, etc.). Aside from these issues, there are also user-induced degradation, as the EOL batteries underwent different usages, handlings and operation conditions. Consequently, regeneration routes like solid-state retholithiation, which require knowledge of lithium deficiency, may encounter scalability challenges, whereas those that do not necessitate such information would be more practical. These points also underscore the importance of rapidly adapting direct recycling methods developed for lab-scale chemically/electrochemically delithiated positive electrode active materials to different types of spent battery chemistries.

Another challenging aspect deals with the thermal stability of degraded electrode materials. For instance, partially delithiated NMC811 already starts to evolve at temperature lower than 300 °C, causing structural changes from layered to rock salt-type structures.^[38,39] The degradation temperature decreases as the lithium content in $\text{Li}_x\text{Ni}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ decreases. Therefore, both pretreatment (delamination) and regeneration experimental conditions should be adapted, i.e. at a temperature below that of degradation, to preserve the structural stability of the active material. Additionally, each regeneration method is often tailored to a specific chemistry or single family of electrode materials. For instance, post-regeneration annealing treatments for the NMC family are typically conducted at elevated temperatures in air, conditions incompatible with the stability of the carbon coating on LFP particles. Thus, as the development of blended electrodes gains traction, aiming to compensate for individual drawbacks of different active material chemistries, it becomes imperative to develop a direct recycling approach compatible with various active material chemistries.

Finally, with ongoing evolution of battery technologies, the development of new material compositions (Ni-rich compounds or $\text{LiFe}_{0.4}\text{Mn}_{0.6}\text{PO}_4$ in replacement of LFP) and the introduction of new coatings or doping on current materials pose a challenge in adapting recycling methods to accommodate these changes. The electrode material chemistry present in the current batteries may no longer be relevant when these batteries will reach their end-of-life in several years. Therefore, even if the researchers successfully develop efficient direct recycling procedures, technological advancements may render these recycled raw materials non-useful in the battery domain. To deal with this issue, the upcycling approach emerges to bring up to date the old active materials. For instance, transforming NMC111 into NMC622 or NMC811, or applying coatings and dopants to degraded materials during their regeneration can improve their performance and extend their use.^[5,40,41] These multi-step processes reported in the literature usually employed a final high temperature annealing in which the degraded material is used as a reactant for the synthesis of up to date active material. This innovative research area shows promise; however once again it seems chemistry-dependent, requires characterization of degraded material, necessitates

several steps that make the recycling less "direct" and may be difficult to adapt to blended active material electrodes.

Directly recycling the negative electrode material, specifically graphite, the most commonly utilized anode material in LIBs, has been less extensively investigated compared to the positive electrode. This is primarily attributed to its economical nature and the limited financial incentive associated with its recycling, even if natural graphite is currently considered as a critical raw material by the European Commission. Regenerating graphite poses a challenge, necessitating the thorough removal of various impurities present on the material's surface and the need to address internal disorder induced by stress resulting from graphite volume expansion during lithium intercalation and potential solvent co-intercalation.^[42] These surface impurities include residual Li, the SEI formed during cycling and binders that hinder performance.^[7] The main approach employed for this purpose consists in a hydrometallurgical step to remove impurities followed by a high temperature annealing (>900 °C).^[7] However, several promising approaches, such as flotation to separate graphite from other components in spent batteries, are under study and show promise to enhance separation and recovery yields. Additionally, some works are conducted to use green solvent to clean graphite surface and lower the carbon footprint of the process. Although these processes under development still need to be optimized to reach satisfactory efficiency and competitive costs, direct recycling of graphite is promising and can help to face the increasing demand of anode material or the EV market. Some approaches developed at lab scale allowed to recover graphite that delivers excellent energy storage performance, comparable to pristine material.^[43,44]

Importance of Life Cycle Assessment

To guide the development of direct recycling processes and to verify their environmental relevance in comparison with primary production and other recycling techniques, comprehensive tools are essential for quantifying environmental impacts. In this context, Life Cycle Assessment (LCA) – a standardized methodology (ISO 14040-44) – proves invaluable in systematically quantifying inputs and outputs related to a system and evaluating associated environmental impacts.^[45,46] It has demonstrated its effectiveness in assessing lithium-ion batteries value chain, including their recycling processes.^[47,48]

Li et al. and Wagner-Wenz et al. reviewed different environmental analysis studies on the recycling of LIBs, assessing the environmental impacts of the different processes.^[49,50] The reviews highlighted challenges in evaluating environmental impacts due to insufficient data for laboratory and industrial processes, depending on the Technology Readiness Level (TRL). Based on the carbon footprint, it suggested that combined hydro-pyrometallurgical processes and direct recycling processes have lower impacts compared to conventional pyrometallurgical processes that require high energy inputs.

However, there is limited research focusing on assessing the potential environmental impacts of innovative direct recycling

processes for LIBs, still under development at lab-scale. In this context, LCA can still be employed to optimize emerging technologies – including direct recycling processes – by identifying environmental hotspots and helping to take eco-design actions, such as solvent selection and managing the trade-off between energy use and process yield.^[51]

Addressing the challenges associated with modelling and upscaling data on direct recycling processes to benchmark with existing technologies involves considering different factors. Firstly, there is a lack of industrial data for separation, delamination and washing processes for direct recycling. Then, the different outputs of the recycling processes, including alloys, metal salts, active materials – for pyro-, hydrometallurgical and direct processes, respectively – further complicate the comparison between these processes. Lastly, the multifunctionality of modelling a recycling process adds another level of complexity. This multifunctionality refers to processes providing more than one function (e.g. treating a waste and obtaining a functional material at the end to reuse).^[52] Direct recycling is expected to outperform existing recycling processes due to lower temperatures and the recovery of higher-value materials (metals oxide instead of metal salts in hydrometallurgical processes) but purification and regeneration stages are introduced especially for EOL. These stages should necessitate low energy and chemicals demand to minimize environmental impacts in comparison with other EOL solutions.

Lastly, at the regulatory level, this holds paramount significance due to the European regulation governing batteries and spent batteries, which mandates the inclusion of carbon footprint assessments throughout the entire life cycle of LIBs.^[2] This encompasses stages from raw materials, manufacturing, and transport to user engagement, and concludes with EOL management (e.g. landfill, recycling) with recovery targets.

Conclusions

LIB direct recycling, also known as “closed-loop recycling” or “electrode materials direct reuse,” is considered as an innovative approach that helps minimize waste, reduce the environmental impact of battery production, and promote a more circular economy in the field of battery. Although a closed loop is achievable, there is no ideal technology that is capable of processing both scraps and EOL batteries. Direct recycling is particularly well-suited for handling electrode production scraps, where active materials are still in pristine state, which does not require exhaustive pretreatments to recover the electrode foils. Physico-chemical delamination seems to emerge as a more promising technique than mechanical or thermal methods, as it allows efficiently removing the binder at low temperatures while preserving the active material integrity. On the other hand, direct recycling will be faced with difficulties with EOL batteries especially if these batteries contain mixed cathodes, on top of the technological advancements and changes in cathode materials. For the active materials containing strategic and valuable elements such as Co or Ni, the hydrometallurgy route which circumvents these issues, will be

difficult to dislodge. Conversely, direct recycling shows promises for LFP chemistry and graphite material, potentially emerging as the sole viable approach to enhance EOL battery recovery yield with cost-effectiveness and a minimized environmental footprint. There is also a need to conduct pilot plant or medium scale tests for practical evaluation of the technology and to facilitate smooth transition to industrial standards. This highlights the crucial need for life cycle assessment analyses to thoroughly evaluate newly designed processes, assessing their impact and pinpointing key areas (hotspots) for improvement. By employing LCA in the direct recycling of LIBs, informed decisions can be taken, optimization of processes, and contribution to a more sustainable energy storage system can be achieved.

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Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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