

DIRECT REGENERATION OF CATHODE MATERIALS IN LITHIUM-ION BATTERIES: THEORETICAL ADVANCES AND TECHNOLOGICAL CHALLENGES

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ABSTRACT: The growing demand for lithium-ion batteries (LIBs), driven by the advancement of electromobility and renewable energy storage, has intensified the need for sustainable strategies for the repurposing of their critical components. Among the recycling methods, the direct regeneration of cathode materials emerges as a promising alternative because it allows the reuse of assets with minimal structural degradation, avoiding dissolution and reprecipitation stages typical of pyrometallurgical and hydrometallurgical routes. The objective of this study is to consolidate the technical-scientific foundations and critically discuss the advances, limitations and operational challenges of direct cathode regeneration in LIBs. This article presents a theoretical review of the physicochemical mechanisms involved, the influence of pretreatment routes and the parameters that affect the efficiency of the process, such as crystal structure, degree of degradation and control of stoichiometry. The main technological barriers to its adoption on an industrial scale are also discussed, including the compositional variability of waste, contamination risks and limitations in the reuse of different cathode families (LCO, NMC, LFP). Finally, the article highlights future perspectives and recommendations for advancing this route based on data from the recent literature, proposing ways for its integration into a circular economy model.

KEYWORDS: Lithium-ion batteries, direct regeneration, cathode recycling, circular economy, sustainability.

1. INTRODUCTION

The increasing insertion of lithium-ion batteries (LIBs) in applications such as electric vehicles, portable electronic devices, and energy storage systems has caused a significant increase in the generation of electrochemical waste at the end of the useful life of these devices. This reality demands the development of technological routes that enable the efficient reuse of the active materials present in LIBs, especially cathode components, whose manufacture represents a significant portion of the cost and environmental impact of the battery life cycle (Fan et al., 2020; Abdalla et al., 2023).

Traditionally, recycling processes are classified into pyrometallurgical routes, hydrometallurgical routes, and, more recently, direct regeneration routes (Georgi-Maschler et al., 2012; Heelan et al., 2016; Fan et al., 2020). Pyrometallurgical approaches involve melting the materials at high temperatures, which simplifies industrial operation, but results in high loss of non-metallic elements, intensive energy consumption, and gaseous emissions (Lv et al., 2018). Hydrometallurgical processes, on the other hand, use aqueous solutions for the selective leaching of metals, allowing high recovery efficiency and purity, but require multiple chemical steps, use of aggressive reagents, and subsequent treatment of effluents (Lai et al., 2021; Ding et al., 2024).

In contrast to these conventional routes, direct regeneration of cathode materials has been investigated as a promising alternative, capable of directly restoring the structure and composition of active materials without the need for total decomposition into their constituent elements (Heelan et al., 2016; Fan et al., 2020). This approach seeks to minimize energy consumption, reduce procedural complexity, and preserve the integrity of materials, which can result in substantial economic and environmental gains (Steward et al., 2019; Abdalla et al., 2023).

Despite the potential, direct regeneration still faces considerable technological and operational challenges, such as the compositional variability of cathode residues, cross-contamination by electrolytes and inert materials, and the difficulty of controlling the stoichiometry of regenerated compounds (Latini et al., 2022; Ding et al., 2024). In addition, efficient reuse depends on the degree of structural degradation of materials after use, requiring rigorous characterization and pretreatment strategies (Lai et al., 2021; Abdalla et al., 2023).

In view of this scenario, this article performs an in-depth theoretical review on the direct regeneration of cathode materials in LIBs, addressing its technical-scientific foundations, the treatment routes involved, the critical parameters of the process and the current limitations for its large-scale application. Through the critical analysis of recent literature, it seeks to consolidate existing knowledge and indicate perspectives for advancement for this technology within a model of circular economy and energy sustainability.

2. THEORETICAL FOUNDATION

2.1 Structure and Composition of Lithium-Ion Batteries

Lithium-ion batteries (LIBs) are electrochemical systems with high energy density, composed fundamentally of an anode, a cathode, an electrolyte and a separator. The anode is generally composed of graphite, while the cathode is formed by lithium-containing transition metal oxides, such as lithium cobalt oxide (LiCoO₂), manganese-nickel-cobalt oxide (NMC), aluminum-nickel-cobalt oxide (NCA), and lithium iron phosphate (LFP) (Abdalla et al., 2023; Tian et al., 2024).

The electrolyte acts as a conductive medium for lithium ions, usually composed of lithium salts (such as LiPF₆) dissolved in volatile organic solvents, while the separator — typically polymers such as polyethylene or polypropylene — prevents direct contact between the electrodes, maintaining the integrity of the system (Lv et al., 2018; Latini et al., 2022). The electrochemical performance of LIB depends directly on the composition and structure of the active materials, especially cathodes, which are responsible for more than 40% of the total cost of the cell (Fan et al., 2020).

Among cathode materials, NMC stands out for its high specific capacity and energy density, while LFP is valued for its thermal stability and safety, although it has a lower energy density (Abdalla et al., 2023). The diversity of cathode compositions and architectures directly influences the regeneration or recycling strategy to be adopted.

2.2 Principles of Direct Regeneration

Direct regeneration consists of restoring the worn cathode material to its original functional state, without the need to chemically decompose the compounds into their constituent elements, as occurs in hydrometallurgical or pyrometallurgical routes (Georgi-Maschler et al., 2012; Fan et al., 2020). This approach aims to maintain the crystal structure and electrochemical properties of the material, minimizing losses and intermediate remanufacturing steps.

The process involves the removal of residual contaminants (such as electrolytes and binders), the adjustment of the stoichiometry of the active elements (usually via relitiation), and controlled heat treatments that restore the crystal structure of the metal oxide (Heelan et al., 2016; Abdalla et al., 2023). Techniques such as assisted co-precipitation, sol-gel, sintering, and sputtering are often employed at different stages of the process (Fan et al., 2020).

The efficiency of direct regeneration depends on factors such as the degree of cell degradation, the uniformity of cathode composition, and the presence of organic or metallic impurities. In addition,

the feasibility of the process is sensitive to the purity of the initial material, requiring rigorous sorting of the waste to be regenerated (Ding et al., 2024; Latini et al., 2022).

2.3 Technical Advantages and Limitations

Among the main advantages of direct regeneration, the reduction in energy consumption and the use of aggressive reagents, the maintenance of the original electrochemical properties of the cathode, and the potential reduction of costs associated with the remanufacture of the material (Steward et al., 2019; Lai et al., 2021). Studies indicate that direct regeneration can present up to 60% energy savings compared to conventional hydrometallurgical routes (Fan et al., 2020).

However, the technology still faces limitations, especially regarding its applicability to different families of cathodes and the difficulty of standardizing post-use waste, which present significant variations in composition, state of degradation, and contamination (Latini et al., 2022; Abdalla et al., 2023). In addition, the absence of widely validated industrial protocols makes direct regeneration a solution still in the pre-commercial stage.

3. TECHNICAL ANALYSIS OF DIRECT REGENERATION ROUTES

The direct regeneration of cathode materials seeks to restore the original electrochemical properties of the active material through routes that preserve its crystal structure, unlike conventional routes based on complete separation of the constituent elements. The main approaches used, their steps, critical parameters and technical challenges are presented below.

3.1 General Steps of the Direct Regeneration Process

According to Fan et al. (2020) and Abdalla et al. (2023), direct regeneration processes generally follow four main steps, as illustrated in Flowchart 1. These steps involve the physicochemical pretreatment of cathode waste, the replacement of lithium stoichiometry, the thermal recrystallization of the active material and, finally, the reconfiguration of the regenerated powder for industrial application. Key steps in the process of direct regeneration of cathode materials from lithium-ion batteries at the end of their useful life. Process efficiency depends on proper residue characterization, control of lithium stoichiometry, and controlled restoration of crystal structure by sintering. Adapted from Fan et al. (2020), Lai et al. (2021), and Abdalla et al. (2023).

Figure 1. Key steps of direct cathode regeneration in lithium-ion batteries.



3.2 Direct Regeneration Techniques Applied to Different Cathodes

3.2.1 NMC Cathodes ($\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$)

NMC materials, due to their high capacity and predominance in the automotive market, have been the most studied in direct regeneration. Studies show that relocation can be performed by impregnation with compounds such as Li_2CO_3 or LiOH , followed by heat treatment (Heelan et al., 2016; Fan et al., 2020).

The main challenge in CMN regeneration is to maintain the proper ratio between transition metals (Ni, Mn, Co), as selective losses can occur during use or pre-treatment (Georgi-Maschler et al., 2012). In addition, the formation of spinel phases or metal segregation can compromise the performance of the regenerated material.

3.2.2 LFP cathodes (LiFePO_4)

LFP, while more stable and safer, presents specific challenges for direct regeneration. Its olivine structure is less tolerant of stoichiometric deviations, and the efficiency of relitiation depends heavily on the integrity of the crystal lattice (Abdalla et al., 2023).

In addition, the presence of oxidized iron (Fe^{3+}) can make it difficult to reverse the ideal redox state, requiring treatments with a reducing atmosphere and precise temperature control (Ding et al., 2024). Doping with conductive carbons or coating with inert materials have also been investigated as ways to recover electrical conductivity and structural stability.

3.2.3 LCO Cathodes (LiCoO_2)

Although less and less used in large-scale applications, LiCoO_2 is still common in portable electronics. Its direct regeneration is relatively well documented, with a high success rate in lithium replacement and recrystallization (Fan et al., 2020).

However, the high cost of cobalt and concerns about environmental toxicity limit its large-scale reuse. Effective separation of impurities, such as organic residues and electrolytes, is essential to avoid contaminations that inhibit crystalline reversal during regeneration (Lai et al., 2021).

3.3 Critical Factors for Direct Regeneration Success

- Composition of the residuals: the variability of the LIBs collected makes it difficult to standardize the operational parameters (Latini et al., 2022);
- Degree of degradation: intense cycling can cause irreversible changes in the crystal structure, making regeneration unfeasible (Fan et al., 2020);
- Cross-contamination: electrolytes, separators, and residual binders can negatively interfere with relitiation and sintering;
- Compatibility with industrial processes: the scalability of direct regeneration depends on adapting the process to automated lines and efficient waste sorting (Steward et al., 2019).

3.4 Benchmarking Other Routes

Table 1 summarizes the main advantages and limitations of direct regeneration compared to hydrometallurgical and pyrometallurgical routes, as per data from Fan et al. (2020), Lai et al. (2021) and Abdalla et al. (2023):

Tabela 1. Summarizes the main advantages and limitations of direct regeneration compared to hydrometallurgical and pyrometallurgical routes

Criterion	Pyrometallurgy	Hydrometallurgy	Direct Regeneration
Energy consumed	High	Moderate	Low
Product purity	Medium	High	High (residual-dependent)
Process steps	Short	Multiple	Multiple
Gas emissions	High	Low	Minimum
Preservation of the structure	Does not preserve	Does not preserve	Preserves
Estimated cost per kg	Medium	High	High
Complexity of scale	Low	High	High

4. DISCUSSION

The theoretical analysis of direct regeneration routes highlights their strategic potential in structuring a circular economy for lithium-ion batteries, especially due to the possibility of reducing costs and environmental impacts. However, this approach is still in the pre-industrial phase, requiring significant advances in process standardization, waste quality control, and adaptation to different families of cathode materials.

As discussed by Fan et al. (2020) and Abdalla et al. (2023), the efficiency of regeneration is strongly conditioned to the stoichiometric control of the regenerated material, especially in the case of nickel-rich cathodes, such as NMC811, which have greater thermal instability and greater sensitivity to degradation of the crystal structure. This condition requires selective lithium reinsertion technologies, controlled doping, and highly accurate heat treatments, factors that still face scalability and reproducibility challenges.

In addition, as highlighted by Latini et al. (2022) and Lai et al. (2021), the lack of uniformity in post-consumer waste imposes a technical-operational bottleneck. The collection of batteries with different compositions, degrees of degradation and contamination requires robust sorting systems, state of health diagnosis (SOH) and efficient segregation routes. Without this, there is a risk of compromising the regeneration and reincorporation of these materials into the production cycle.

In the case of LFP cathodes, direct regeneration encounters additional obstacles. The olivine structure has lower tolerance to cationic disorder, and lithium reinsertion kinetics are less efficient than in NMC or LCO systems (Ding et al., 2024). Thus, although LFP offers thermal safety and stability in operation, its direct reuse still requires further investigation.

From an environmental and energy point of view, the benefits of direct regeneration are evident. Studies by Fan et al. (2020) indicate that this approach can reduce energy consumption per kilogram of regenerated material by up to 70% compared to pyrometallurgical routes, in addition to minimizing emissions of harmful gases and the generation of chemical effluents. However, as pointed out by

Steward et al. (2019), the economic viability of the technology still depends on external factors such as environmental regulation, recycling subsidies, and the development of collection infrastructure.

Despite the challenges, recent experiences point to concrete advances. The ReCell Center (USA), an initiative led by the United States Department of Energy (DOE), has excelled in the development of direct regeneration routes with a focus on NMC and LCO, using controlled lithium reinsertion routes and repurposing of crystalline structures without the need for total refabrication (Fan et al., 2020). Similarly, companies such as Battery Resources (now Ascend Elements) have developed commercial solutions based on direct regeneration, aimed at producing ready-to-use cathodes from post-consumer waste, with purity rates comparable to primary materials.

These cases demonstrate that direct regeneration is not just a conceptual proposal, but an expanding technological route, whose feasibility depends on integration with automated industrial processes, intelligent waste sorting and strict control of processing conditions.

Thus, although direct regeneration does not completely replace other recycling routes in the short term, it presents itself as an essential complementary pillar, especially for high value-added materials such as NMC cathodes. Integrating this route into a hybrid recycling chain, combining automated mechanical separation, sensor diagnostics, and thermal-selective regeneration, may represent the most viable path for its future industrial adoption.

5. FINAL CONSIDERATIONS

The direct regeneration of cathode materials in lithium-ion batteries emerges as a technologically promising and environmentally strategic alternative in view of the limitations of conventional recycling routes. By allowing the recovery of active materials without complete dissolution or melting, this approach preserves the crystal structure of the compounds, reduces energy consumption, minimizes emissions, and avoids the generation of aggressive chemical effluents.

The theoretical analysis presented in this work demonstrates that, although the scientific foundations of direct regeneration are well established for certain cathode families, such as LCO and NMC, its consolidation as a viable solution on an industrial scale still requires the confrontation of relevant challenges. Among the main obstacles are the compositional heterogeneity of the waste, the need for strict control of the stoichiometry during the lithium reinsertion process and the operational limitations associated with the variability of the degradation state of the post-use materials.

On the other hand, the potential gains, economic, environmental and operational, justify the investment in applied research, development of hybrid routes and advances in sorting, characterization and selective heat treatment technologies. In addition, the creation of specific regulations and incentives for the efficient recycling of LIBs can accelerate the industrial adoption of this route, favoring the transition to circular and sustainable production models.

It is therefore concluded that direct regeneration should not be seen as an immediate substitute for the pyrometallurgical and hydrometallurgical routes, but as a complementary technology with high added value, especially in contexts in which the recovery of functional materials represents a technical and economic advantage. Future research should focus on the development of standardized selective lithium reinsertion protocols, the use of automated screening techniques, and the efficient repurposing of mixed cathodes, with the aim of increasing the robustness and standardization of the process on an industrial scale.

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