

RECYCLING LITHIUM-ION BATTERIES: A STUDY OF RECYCLING METHODS

LEONARDO HENRIQUE GOMES¹, MARIA JOSÉ J. DE S. PONTE², HAROLDO DE A. PONTE³, HENRIQUE DE SANTANA⁴ e GABRIEL HENRIQUE GOMES⁵

¹Department of Mechanical Engineering, Federal University of Paraná (UFPR). Curitiba-PR, leonardohgomes.ufpr@gmail.com

²Department of Mechanical Engineering, Federal University of Paraná (UFPR). Curitiba-PR

³Department of Chemical Engineering, Federal University of Paraná (UFPR). Curitiba-PR

⁴Department of Chemistry, Bioenergy Program, State University of Londrina (UEL). Londrina-PR

⁵Civil Engineering Technical Scholarship, Projetek, Araucária Foundation, Jacarezinho-PR

Presented at Technical Scientific Congress of Engineering and Agronomy - CONTECC October 07-10, 2024

ABSTRACT: The recycling of lithium-ion batteries has become essential due to the growing demand for energy in electronic devices and electric vehicles, which increases the presence of these batteries on the global market. With the increase in these discarded batteries, new opportunities arise for the recovery and recycling of valuable components, such as cathodes and anodes, which can be reused in the production of new batteries. This article reviews the methods of treating spent lithium-ion battery cathode materials, looking at various techniques including mechanical, pyrometallurgical, hydrometallurgical and electrochemical processes. Standard recycling approaches are grouped into three main categories: pyrometallurgy, hydrometallurgy and direct recycling. In addition to reviewing the processes, the article analyzes the economic and environmental challenges associated with their recycling and discusses the future prospects and applications of these methods. Continued research and the development of new technologies are crucial to improving the efficiency and sustainability of recycling, meeting the growing demands for more effective solutions in the management of waste batteries.

KEYWORDS: Lithium-ion batteries, treatment processes, recovery of metals and components, economic and environmental challenges.

1. INTRODUCTION

According to Sun et al. (2017) and Lai et al. (2021), due to intensive research into lithium-ion batteries, it has been identified that they have significant advantages over other types of energy storage devices. According to Tian et al. (2024), lithium-ion batteries (LIBs) are considered an advanced energy storage technology, playing a key role in renewable and sustainable electrification. According to Abdalla et al. (2023), with high energy and power density, as well as a long life cycle, these batteries are essential for the operation of electronic devices and electric vehicles.

According to Latini et al. (2022) and Ding et al. (2024), separators, important components within a LIB, keep anodes and cathodes mechanically separated, allowing for maximum ionic conductivity of lithiumion electrolytes. According to Abdalla et al. (2023) and Sun et al. (2017), the design and performance of LIBs directly affect battery capacity, life cycle and safety. According to Lai et al. (2021), worn-out battery packs are often removed from electric vehicles and discharged before being divided into individual modules or cells, because according to Tian et al. (2024), as the state of health of these batteries is usually still between 70% and 80% of the initial capacity, many can be reused in a second life, such as stationary energy storage in homes. The reuse of recycled LIBs in buildings and public services can result in cost savings of 13-57% and a reduction in greenhouse gas emissions of 7-32%.

According to Latini et al. (2022), if the module's state of health is lower, it can be discharged, recycled or dismantled and reconfigured at the cell level. In such cases, cells with low state of health (SOH) values are recharged to extend the module's useful life. According to Steward et al. (2019) and Fan et al. (2020), recycling second-hand LIBs consumes less energy and has a smaller carbon footprint.





However, whether they come from electric vehicles or secondary use, LIBs eventually need to be recycled, making recycling essential for a closed-loop economy.

This article explores the standard approach to recycling LIBs, which involves a variety of mechanical, physical, thermal and chemical treatments, grouped into three main categories: pyrometallurgy, hydrometallurgy and direct recycling. Pyrometallurgy, although it has a shorter process chain, involves significant energy use and GHG emissions, as well as material loss. On the other hand, hydrometallurgy uses less energy and emits fewer hazardous gases, providing greater purity of the recovered materials. In both cases, direct recycling requires dismantling the cells down to the individual electrodes for efficient separation and collection of the active elements.

This introduction puts into context the importance of LIBs and the need to develop efficient recycling methods to ensure sustainability and minimize the environmental impacts associated with their disposal.

2. LITERATURE REVIEW

2.1 Composition, Performance and Sustainability of Lithium-Ion Batteries (LIBs)

According to Latini et al. (2022), the literature review on lithium-ion batteries (LIBs) addresses the composition, performance and sustainability of these essential technologies for portable electronic devices and electric vehicles. In terms of chemical composition, LIBs are made up of several key elements. The anode, usually made of graphite, allows lithium ions to intercalate during charging and to be released during discharge. Research is underway to replace graphite with silicon, which would provide greater capacity, although this material faces challenges related to volumetric expansion. Abdalla et al. (2023) and Tian et al. (2024) state that the cathode is typically made up of materials such as lithium cobalt oxide (LiCoO₂) and lithium iron phosphate (LiFePO₄). Lithium-nickel-manganesecobalt oxide (NMC) is an alternative that offers high capacity and energy density, while LFP (lithiumiron phosphate) is recognized for its stability and safety. According to Fan et al. (2020), the electrolyte consists of a lithium salt solution dissolved in an organic solvent, facilitating the movement of ions between the anode and cathode. Recently, solid electrolytes have been developed with the aim of increasing safety. LV et al. (2018) indicate that the separator, which keeps the anode and cathode isolated, is usually made of polymers such as polyethylene or polypropylene, preventing short circuits. There is also growing interest in separators that have automatic erasing properties, thus increasing battery safety. Finally, Sun et al. (2017) mention that current collectors, made up of copper foil for the anode and aluminum foil for the cathode, are responsible for conducting the electric current to the external circuit.

Georgi-Maschler et al. (2012) and Steward et al. (2019) point out that, in terms of performance, the capacity of LIBs is strongly influenced by the materials used in the anode and cathode. Graphite, widely used in the anode, can be replaced by silicon to achieve a higher capacity, while in the cathode, materials such as NMC and LFP offer different balances between capacity, stability and safety. Performance optimization is achieved by choosing materials that promote the efficient intercalation of lithium ions and reduce internal resistance, which results in greater energy efficiency and better charging and discharging rates.

Heelan et al. (2016) and Xu et al. (2008) note that in terms of sustainability, LIBs have played a crucial role in reducing greenhouse gas emissions and promoting a transition to a cleaner energy economy. However, Ding et al. (2024) add that the environmental challenges are significant, as the infrastructure for recycling and disposing of batteries is still insufficient. This leads to concerns about the environmental and safety impacts associated with the improper handling of batteries after use. Lai et al. (2021) state that research and the development of new technologies seek to improve the sustainability of LIBs, with efforts focused on reducing costs and lowering environmental impact.

Abdalla et al. (2023) add that future innovations include the use of safer and more sustainable materials, such as solid electrolytes and cobalt-free cathodes, with the aim of reducing the environmental footprint throughout the life cycle of the batteries.

3. RECYCLING METHODS





According to LV et al. (2018) and Fan et al. (2020), recycling methods for lithium-ion batteries (LIBs) seek to isolate and recover valuable resources, such as metals and other components, after the batteries have reached the end of their useful life. According to Abdalla et al. (2023), the recycling process can be divided into several stages and techniques, each with its own advantages and challenges.

According to Lai et al. (2021) and Steward et al. (2019), recycling lithium-ion batteries (LIBs) involves several techniques, each with its own characteristics and challenges. Thermal separation uses high temperatures to decompose and separate battery components and is effective in recovering valuable metals, but can result in material losses and harmful gas emissions.

Hydrometallurgy uses aqueous solutions to dissolve the desired metals, allowing for clean and efficient recovery with lower energy consumption compared to thermal processes. However, the effectiveness of this method depends on specific conditions, such as the solid-liquid ratio and temperature.

Biohydrometallurgy, an innovative approach, uses living organisms, such as bacteria or fungi, to dissolve the electrode materials. This method has the potential for high efficiency when using pure acids, but faces challenges related to pulp density and iron and sulphur contamination.

Direct regeneration involves the recovery and regeneration of anode and cathode materials using techniques such as co-dripping and the sol-gel method. Although it offers high efficiency in regenerating materials, it can be impacted by contamination and the variability of recycled batteries.

According to Georgi-Maschler et al. (2012) and Heelan et al. (2016), combined methods, which integrate techniques such as solvent extraction, precipitation and electrolysis, aim to optimize material recovery. Despite improving efficiency and maximizing recovery, the complexity of the process can increase the cost and time of recycling.

The choice of recycling method for LIBs depends on the type of battery, the materials to be recovered and the specific operating conditions. Each technique has its advantages and challenges, and combining methods can offer a more efficient and sustainable solution for the recovery of battery components.

4. RESULTS AND DISCUSSION

Recycling methods for lithium-ion batteries (LIBs) are essential to ensure the recovery of valuable resources and reduce the environmental impact of the improper disposal of these batteries. Each method has specific advantages and challenges, and it is necessary to select the appropriate technique based on the characteristics of the batteries and the materials to be recovered. Below is a detailed analysis of the main recycling techniques:

Recycling method	Process	Advantages	Disadvantages
Thermal Separation	Uses high temperatures to break down and separate battery components.	 Effective in recovering valuable metals such as cobalt and nickel; Can treat large volumes of material. 	 Loss of valuable material due to volatilization; Toxic gas emissions and the need for strict environmental control.
Hydrometallurgy	It uses aqueous solutions to dissolve the desired metals.	 High efficiency in recovering high purity metals; Lower energy consumption compared to thermal processes; Possibility of selective recovery of specific metals. 	 Production of liquid effluents that require treatment; Dependence on chemical reagents which can be expensive and generate waste.





Biohydrometallurgy	It uses living organisms, such as bacteria or fungi, to dissolve the electrode materials.	 Environmentally friendly method, with less use of aggressive chemical products; Potential for recovering metals in more dilute solutions. 	 Slower processes compared to chemical and thermal methods; Sensitivity to environmental conditions and the presence of contaminants.
Direct regeneration	It directly recycles and synthesizes cathode and anode materials.	 High efficiency in the recovery and reuse of materials; Possibility of regenerating graphite and cathode materials with properties comparable to the originals. 	 Contamination of regenerated materials can affect efficiency; Variability in the composition of recycled batteries can complicate the process.
Combined Methods	It integrates various techniques to optimize material recovery.	 Combination of methods maximizes recovery and process efficiency; Flexibility to adjust the process according to the specific needs of the materials to be recovered. 	 More complex and potentially more expensive processes; Requires an in-depth understanding of the interactions between the different techniques.

The choice of recycling method depends on several factors, such as the type of battery, as different compositions of lithium-ion batteries (LIBs) may require different approaches. In addition, the materials to be recovered are a crucial factor, as some methods are more efficient for recovering certain metals. Operating conditions also play an important role, including available infrastructure, operating costs and environmental regulations.

Combining techniques can offer more robust and sustainable solutions, allowing the recovery of the valuable resources present in lithium-ion batteries to be maximized. Continuous research and the development of new technologies are essential to improve recycling processes and meet the growing demands for sustainability and efficiency in the management of waste batteries.

5. CONCLUSION

Intensive research into lithium-ion batteries (LIBs) has revealed their significant advantages as advanced energy storage devices, key to renewable and sustainable electrification. With their high energy density, power and long service life, LIBs are essential for electronic devices and electric vehicles. Separators, critical components within a LIB, ensure mechanical separation between anodes and cathodes, allowing for maximum ionic conductivity of the electrolytes. The importance of the design and performance of LIBs is evident, directly affecting the capacity, life cycle and safety of the battery.

The reuse of recycled LIBs can result in significant cost savings and a reduction in greenhouse gas emissions. When the state of health (SOH) of the batteries is low, they can be discharged, recycled or disassembled and reconfigured. Recycling LIBs consumes less energy and has a smaller carbon footprint. Recycling methods such as pyrometallurgy, hydrometallurgy and direct recycling are essential for a closed-loop economy, ensuring the efficient recovery of valuable materials and minimizing environmental impacts.

In conclusion, continued advancement in LIB recycling techniques is crucial to sustaining the growing demand for energy, while promoting sustainability and reducing the environmental impact associated with the improper disposal of these batteries.

ACKNOWLEDGMENTS

This study was funded by the Coordination for the Improvement of Higher Education Personnel - Brazil (CAPES) - Code 001. Partner institutions: Department of Mechanical Engineering and Department of Chemical Engineering at the Federal University of Paraná (UFPR). PhD Program in Bioenergy at the Chemistry Department of the State University of Londrina (UEL).





ABDALLA AM, ABDULLAH MF, DAWOOD MK, WEI B, SUBRAMANIAN Y, AZAD AT, et al. **Innovative lithium-ion battery recycling: Sustainable process for recovery of critical materials from lithium-ion batteries.** J Energy Storage. 2023;67:107551. doi:10.1016/j.est.2023.107551. Available from: <u>https://www.sciencedirect.com/science/article/pii/S2352152X23009489</u>. Accessed: 02 Aug 2024.

DING, G.; LIU, F.; FAN, X.; GAO, X.; CAO, G.; BAN, J.; LI, Z.; HU, J. **Research on green** recycling of lithium-ion batteries cathode waste powder. *Chemical Engineering Journal*, v. 493, p. 152837, 2024. ISSN 1385-8947. doi:10.1016/j.cej.2024.152837. Available from: <u>https://www.sciencedirect.com/science/article/pii/S1385894724043249</u>. Accessed: 02 Aug 2024.

FAN E, LI L, WANG Z, LIN J, HUANG Y, YAO Y, et al. Sustainable recycling technology forlithium-ionbatteriesandbeyond:Challengesandfutureperspectives.ChemRev.2020;120(14):7020-63.doi:10.1021/acs.chemrev.9b00535.Availablefrom:https://pubs.acs.org/doi/10.1021/acs.chemrev.9b00535.Accessed: 02 Aug 2024.

GEORGI-MASCHLER T, FRIEDRICH B, WEYHE R, HEEGN H, RUTZ M. Development of a recycling process for Li-ion batteries. J Power Sources. 2012;207:173-82. doi:10.1016/j.jpowsour.2012.01.152. Available from: https://www.sciencedirect.com/science/article/pii/S0378775312002984. Accessed: 02 Aug 2024.

HEELAN J, GRATZ E, ZHENG Z, et al. Current and prospective processes for recycling and recovery of lithium-ion batteries. JOM. 2016;68:2632-8. doi:10.1007/s11837-016-1994-y. Available from: https://doi.org/10.1007/s11837-016-1994-y. Accessed: 02 Aug 2024.

LAI X, HUANG Y, GU H, DENG C, HAN X, FENG X, ZHENG Y. **Turning waste into wealth: A systematic review on echelon utilization and material recycling of retired lithium-ion batteries**. Energy Storage Mater. 2021;40:96-123. doi:10.1016/j.ensm.2021.05.010. Available from: https://www.sciencedirect.com/science/article/pii/S2405829721002142</u>. Accessed: 02 Aug 2024.

LATINI D, VACCARO M, LAGNONI M, OREFICE M, MATHIEUX F, HUISMAN J, et al. A comprehensive review and classification of unit operations with assessment of outputs quality in lithium-ion battery recycling. J Power Sources. 2022;546:231979. doi:10.1016/j.jpowsour.2022.231979. Available from: https://www.sciencedirect.com/science/article/pii/S0378775322009594. Accessed: 02 Aug 2024.

LV W, WANG Z, CAO H, SUN Y, ZHANG Y, SUN Z. A critical review and analysis of the recycling of used lithium-ion batteries. ACS Sustain Chem Eng. 2018;6(2):1504-21.

from:

doi:10.1021/acssuschemeng.7b03811. Available

https://pubs.acs.org/doi/10.1021/acssuschemeng.7b03811. Accessed: 02 Aug 2024.

STEWARD D, MAYYAS A, MANN M. Economics and challenges of Li-ion battery recycling from end-of-life vehicles. Procedia Manuf. 2019;33:272-9. doi:10.1016/j.promfg.2019.04.033. Available from: https://www.sciencedirect.com/science/article/pii/S2351978919305104. Accessed: 02 Aug 2024. SUN X, HAO H, ZHAO F, LIU Z. Tracing global lithium flow: A trade-linked material flow analysis. Resour Conserv Recycl. 2017;124:50-61. doi:10.1016/j.resconrec.2017.04.012. Available from: https://www.sciencedirect.com/science/article/abs/pii/S0921344917301118. Accessed: 02 Aug 2024.

TIAN, S.; CAO, Y.; DONG, L.; SHI, P.; LI, R.; JIN, Y.; TU, Y.; DU, C.; REN, Z.; ZHOU, Z. Selective recovery of lithium from spent LiFePO₄ powders with electrochemical method. *Journal of Environmental Chemical Engineering*, v. 12, n. 3, p. 112871, 2024. ISSN 2213-3437. doi:10.1016/j.jece.2024.112871. Available from: https://www.sciencedirect.com/science/article/pii/S2213343724010017. Accessed: 02 Aug 2024.

XU J, THOMAS HR, FRANCIS RW, LUM KR, WANG J, LIANG B. A review of processes and technologies for the recycling of lithium-ion secondary batteries. J Power Sources. 2008;177(2):512-27. doi:10.1016/j.jpowsour.2007.11.074. Available from: https://www.sciencedirect.com/science/article/pii/S0378775307026195. Accessed: 02 Aug 2024.

