

Sustainable Lithium-Ion Battery with Environmental Assessment: From Mine to Battery-Grade Salts

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Abstract

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Electric vehicles (EVs) play a pivotal role in decarbonizing transportation by providing a cleaner alternative to fossil fuels. However, realizing their full sustainability potential requires the entire battery supply chain to be environmentally friendly. Lithium-ion batteries have enabled the rapid adoption of EVs, yet life cycle assessments (LCAs) show that their highest environmental impact stems from upstream raw material production. This thesis addresses these challenges by examining electrode components through three perspectives: lithium production, nickel extraction, and binder sustainability. First, we conducted an LCA of $\text{LiOH} \cdot \text{H}_2\text{O}$ production in Québec. The results indicate a short-term climate change impact of 4.395 t $\text{CO}_2\text{-eq}$ per ton of $\text{LiOH} \cdot \text{H}_2\text{O}$ and a water scarcity footprint of 9,422.31 m^3 world eq after using sodium sulfate co-products credits. Natural gas combustion in the calciner was identified as the primary contributor to emissions, highlighting electrification of the process as a promising mitigation pathway given Québec's hydroelectric energy mix. Next, the thesis explores nickel recovery through the sulfation-roasting process using a dual-salt additive system of Na_2SO_4 and K_2SO_4 . As Na_2SO_4 is a by-product from $\text{LiOH} \cdot \text{H}_2\text{O}$ production, its usage for nickel recovery promotes a circular economy within critical mineral supply chains. Under optimized conditions (650 °C, 2.5 h, 4 g Na_2SO_4 , 1.25 g K_2SO_4), the process achieved 79% Ni extraction and 95% Fe recovery, with the recovered iron oxide suitable as a precursor for LiFePO_4 cathode materials. Finally, the thesis evaluates sustainable aqueous binders as replacements for PVDF/NMP systems, addressing toxicity and solvent-related impacts.

Keywords: critical minerals, transformation from ore to salts, pyro-hydrometallurgy, ore processing

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Contribution of Authors

Chapter 1: Written entirely by the sole author of this thesis. Portions of this chapter are adapted from the published article “*Binders for Li-Ion Battery Technologies and Beyond: A Comprehensive Review*,” for which the thesis author is the first author. The thesis author was responsible for designing the review’s structure, collecting relevant papers, writing the manuscript, and creating the graphics. Co-author Anil Kumar Reddy contributed by gathering papers pertinent to the review topic, while co-author Karim Zaghib focused on reviewing the manuscript and refining its language.

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Chapter 4: Based on published article, “*Life Cycle Assessment of Nickel, Manganese, Cobalt Critical Minerals: Lithium Hydroxide Monohydrate (Mine-to-Material) in Québec, Canada*.” The thesis author contributed to data collection, execution and review of the LCA model in the software and drafting of the manuscript. Author Gary Vegh performed and reviewed the LCA model and drafted the manuscript. Co-authors Sarah Sajedi and Ivan Kantor reviewed both the LCA model and the manuscript. Co-authors Khalil Amine and Anil Kumar Reddy reviewed the manuscript, while Co-author Karim Zaghib provided review and overall supervision of the research and manuscript preparation.

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Chapter 1 - Introduction

1.1. Lithium-ion batteries

The escalating issue of climate change has led to a significant increase in interest in renewable energy sources as viable alternatives to traditional non-renewable resources. The rising risk of surpassing the Intergovernmental Panel on Climate Change's (IPCC) critical 1.5 °C temperature threshold underscores the urgent need for action [1, 2]. Specifically, reducing greenhouse gas (GHG) emissions has become a matter of paramount importance in mitigating the potential consequences of global warming. Recent data highlight the severity of the situation; in 2022, the global concentration of CO₂ reached a concerning level of 417.1 parts per million (ppm), representing a 23% increase in CO₂ levels since 1980 [3]. The major contributing factors to GHG emissions include burning fuels, particularly fossil fuels, for transportation or electricity production. Technological advancements are essential not only for meeting the ever-increasing energy demands of a growing global population but also for overcoming the inherent limitations and environmental impacts associated with the continued reliance on fossil fuels as our primary energy source [4].

Among the various strategies adopted to address climate change, the electrification of society has emerged as a prominent and increasingly influential approach. Electrification involves transitioning from fossil fuel-based technologies to systems that are not only more energy-efficient but also significantly reduce CO₂ emissions. This transformation will play a critical role in achieving the Net Zero Emissions (NZE) targets set for 2050. A central component of this shift is the development and adoption of electric vehicles (EVs), which play a crucial role in decarbonizing the transportation sector, which is responsible for approximately 15% of global GHG emissions. The transportation sector's reliance on fossil fuels makes it a prime target for emissions reduction efforts, and EVs offer a viable and increasingly popular alternative. The momentum behind EV adoption continues to accelerate, with global sales rising from approximately 0.3 million in 2014 to over 17 million in 2024. Projections suggest that this figure will exceed 20 million by 2025 [5]. The global EV market will reach USD 784.2 billion by 2025 and grow at a CAGR of 6.01%, ultimately reaching approximately USD 990.4 billion by 2029 [6]. This steady expansion in EV uptake is a promising indicator of progress toward the NZE 2050 objectives, demonstrating a tangible shift towards a more sustainable transportation ecosystem and a lower-carbon future.

The effective deployment of EVs is fundamentally dependent on advancements in energy storage technologies, particularly battery energy storage systems. Battery demand in EVs will reach 3 TWh by 2030 [7]. The advent of rechargeable batteries not only transformed the consumer electronics industry but also served as a cornerstone in the ongoing evolution of electric mobility [8]. Among the various rechargeable battery technologies, lithium-ion batteries (LIBs) have experienced the most significant progress. Since their commercial introduction by Sony in 1991, the energy density of LIBs has increased substantially, from approximately 80 Wh kg⁻¹ to around 250 Wh kg⁻¹ [9]. Innovations in cathode and anode materials primarily drive the enhancement in energy density. These advancements include modifications to the chemical composition of the electrode materials and design, as well as the strategic application of surface coatings and elemental doping techniques. These combined efforts have collectively led to enhanced structural stability, improved interfacial integrity between the electrode and electrolyte, and superior overall electrochemical performance of the LIBs. Nevertheless, further development is essential to meet the projected energy density demands exceeding 350 Wh kg⁻¹, which are necessary to support the

accelerating adoption of EVs [10]. The pursuit of higher energy density LIBs remains a critical focus for researchers and engineers in the field.

Achieving high energy density in LIBs requires a comprehensive evaluation and optimization of the entire value chain, from sourcing battery-grade raw materials for electrode production to cell and pack manufacturing. Technological advancements are required at each stage of the LIB lifecycle, not only to enhance energy density but also to ensure operational safety, which remains a significant concern. Additionally, the environmental impact of LIBs must be critically assessed, particularly given the positioning of EVs as a cleaner alternative to conventional gasoline-powered vehicles. EVs emit no direct GHGs during use, but their overall environmental footprint depends on mineral extraction, electricity production sources, and battery end-of-life management. Ensuring that these processes are environmentally sustainable is essential to substantiate the role of EVs as a genuinely green technology.

1.2. Scope of work

This thesis aims to critically analyze and develop sustainable processes for LIBs. It integrates a thorough review of existing literature on topics associated with LIBs, along with original experimental and environmental assessment. Advancing clean and sustainable technologies for LIBs, this thesis focuses on two areas: (i) raw material transformation and (ii) evaluation of a key electrode component.

Concerning raw materials, the research emphasizes two critical minerals: nickel and lithium.

- Lithium: Life cycle assessment (LCA) of battery-grade lithium hydroxide production from spodumene ore in Quebec.
- Nickel: Development and optimization of extraction process from sulfide ore using a novel dual-salt additive sulfate roasting–leaching method.

The thesis also critically reviews environmentally friendly alternatives to conventional toxic binders, such as polyvinylidene fluoride (PVDF), widely used in LIB electrodes.

A brief overview of the thesis structure is as follows:

- Chapter 2 discusses the importance of establishing a circular economy framework across the LIB value chain, from raw material extraction to end-of-life recycling.
- Chapter 3 reviews the transformation of lithium and nickel from ores into battery-grade salts, which serve as precursors for electrode materials.
- Chapter 4 presents an environmental assessment of lithium hydroxide monohydrate ($\text{LiOH}\cdot\text{H}_2\text{O}$) production from spodumene ore using LCA methodology.
- Chapter 5 presents the experimental work focused on optimizing nickel extraction from sulfide ore through sulfation-roasting method
- Chapter 6 provides a critical review of binder systems used in LIB electrodes, with a focus on sustainable alternatives to conventional polymeric binders like PVDF.

References

1. P. Pablo-Romero et al., “A Global Assessment: Can Renewable Energy Replace Fossil Fuels by 2050?,” *Sustainability* 2022, Vol. 14, Page 4792, vol. 14, no. 8, p. 4792, Apr. 2022, doi: 10.3390/SU14084792.
2. “Global Warming of 1.5 oC —.” Accessed: Jun. 17, 2025. [Online]. Available: <https://www.ipcc.ch/sr15/>
3. “Greenhouse gas concentrations - Canada.ca.” Accessed: Jun. 17, 2025. [Online]. Available: <https://www.canada.ca/en/environment-climate-change/services/environmental-indicators/greenhouse-gas-concentrations.html>
4. M. Srivastava, A. K. Anil, and K. Zaghib, “Binders for Li-Ion Battery Technologies and Beyond: A Comprehensive Review,” *Batteries* 2024, Vol. 10, Page 268, vol. 10, no. 8, p. 268, Jul. 2024, doi: 10.3390/BATTERIES10080268.
5. “Electrification - Energy System - IEA.” Accessed: Jun. 17, 2025. [Online]. Available: <https://www.iea.org/energy-system/electricity/electrification>
6. “Electric Vehicles - Worldwide | Statista Market Forecast.” Accessed: Jul. 08, 2025. [Online]. Available: <https://www.statista.com/outlook/mmo/electric-vehicles/worldwide>
7. “Electric vehicle batteries – Global EV Outlook 2025 – Analysis - IEA.” Accessed: Jul. 08, 2025. [Online]. Available: <https://www.iea.org/reports/global-ev-outlook-2025/electric-vehicle-batteries>
8. M. Li, J. Lu, Z. Chen, and K. Amine, “30 Years of Lithium-Ion Batteries,” Aug. 16, 2018, Wiley-VCH Verlag. doi: 10.1002/adma.201800561.
9. Y. Nishi, “Lithium ion secondary batteries; past 10 years and the future,” *J Power Sources*, vol. 100, no. 1–2, pp. 101–106, Nov. 2001, doi: 10.1016/S0378-7753(01)00887-4.
10. F. M. N. U. Khan, M. G. Rasul, A. S. M. Sayem, and N. Mandal, “Maximizing energy density of lithium-ion batteries for electric vehicles: A critical review,” *Energy Reports*, vol. 9, pp. 11–21, Oct. 2023, doi: 10.1016/J.EGYR.2023.08.069.

Chapter 2 - Circular Economy Strategies Across LIB Lifecycle

2.1. Introduction to Circular Economy

The Ellen MacArthur Foundation defines circular economy as "a system where materials never become waste and are instead regenerated". The core objective of a circular economy is to maximize the utility of products and materials through strategies such as maintenance, reuse, refurbishment, remanufacturing, recycling, and composting. This approach fundamentally contrasts with the traditional "take-make-waste" or linear economic model, wherein resources are extracted, converted into products, and ultimately discarded after use. The circular economy, on the other hand, offers a more sustainable and resource-efficient alternative. Three key principles guide the circular economy: (a) eliminating waste and pollution, (b) circulating products and materials within the economy, and (c) regenerating natural systems [1].

The circular economy offers several key benefits. First, it makes a significant contribution to environmental protection. By promoting the reuse and recycling of products, the reliance on virgin natural resources is minimized, thereby alleviating environmental pressure. Thus, not only conserves resources but also leads to a measurable reduction in GHG emissions. Furthermore, the circular approach generates minimal to zero waste. Second, it enhances the stability of raw material supply. Recycling reduces the risks associated with raw material procurement, which geopolitical factors often influence. By diversifying supply sources and reducing dependency on primary extraction, the circular economy contributes to a more resilient and stable materials market with reduced price volatility and, lastly, socioeconomic advantages. The transition to a circular model can foster increased competitiveness and drive innovation across industries. Moreover, the production of durable, long-lasting goods can lead to cost savings for consumers, reinforcing economic resilience and sustainability [2].

Recognizing the multiple benefits of a circular economy, regions such as Europe and China have emerged as global leaders in its implementation. In 2020, the European Union adopted the New Circular Economy Action Plan (CEAP), a strategic component of the EU's broader objective to achieve climate neutrality by 2050. CEAP outlines measures aimed at promoting sustainable product design, reducing waste generation, empowering consumers and public purchasers, and targeting high-resource-consuming sectors for reform. Through this initiative, Europe seeks not only to transition domestically but also to position itself as a global frontrunner in circular economy practices [3]. China had institutionalized its commitment to the circular economy much earlier, initiating the 3R principles—Reduce, Reuse, and Recycle—in 2008. Since then, the country has progressively advanced its circular policies, culminating in its 14th Five-Year Plan for Circular Economy Development (2021–2025). This plan sets ambitious targets, including a 20% increase in resource productivity compared to the previous plan, as well as reductions of 13.5% and 16% in energy and water consumption per unit of GDP, respectively, alongside several other resource-efficiency goals [4].

The transition to a circular economy is essential across all sectors. In particular, the growth of EVs and portable electronic devices drives increasing battery demand, highlighting the critical need to implement circular economy principles in the battery industry. By adopting such principles, the battery industry can mitigate environmental impacts, reduce reliance on raw materials, and foster a more sustainable and resilient supply chain.

2.2. Need for circular economy in LIBs

The significant distinction between the linear and circular economic models emerges at the end-of-life (EOL) stage. In a circular system, LIBs are subjected to recycling and reuse, whereas in a linear model, they are typically discarded as waste [5]. As LIB demand continues to rise, the linear model risks generating substantial volumes of battery waste. Given that LIBs generally have a lifespan of 5 to 10 years [6], projections indicate that by 2040, approximately 4 million tons of spent LIBs will be generated annually [7]. This scale of waste poses both environmental and economic challenges, as its management would require significant financial resources. Furthermore, the presence of toxic substances within LIBs poses serious environmental risks; improper disposal of spent LIBs would enter the landfills, which not only threatens ecosystems but also increases the likelihood of fire hazards [8].

LIBs also rely heavily on critical raw materials such as lithium, nickel, cobalt, and manganese, which are elements with finite global reserves. According to the linear economic model, these essential resources could be depleted sooner, potentially hindering the future expansion of electrification. Moreover, the mining and processing of these minerals have considerable environmental implications. For instance, the extraction of lithium from brine has a significant impact on local water availability. At the same time, the chemicals used in leaching and purification can contaminate water bodies and ecosystems if not properly managed. From a social perspective, artisanal cobalt extraction has raised ethical concerns due to reports of human rights violations and exploitative labor practices [5].

The adoption of circular economy strategies offers a pathway to mitigate these issues. By promoting the recovery and reuse of valuable materials, circularity reduces the strain on natural resources, lowers production costs, and generates additional revenue streams [9]. Furthermore, a circular approach enhances the sustainability profile of EVs, reinforcing their role as a genuinely green alternative to conventional transportation.

2.3. Policies

To promote the adoption of circular economy principles in LIBs, several countries have introduced targeted policies aimed at reinforcing their positions in the LIB value chain. EU has officially recognized “black mass”, the material recovered after the pre-treatment of spent batteries, as hazardous waste. This designation aims not only to safeguard the environment from hazardous waste but also to keep black mass within the region. By doing so, the EU aims to enhance the security of raw material supply and establish a robust domestic recycling infrastructure, thereby advancing its circular economy objectives [10]. In the United States, the National Blueprint for Lithium Batteries outlines a comprehensive strategy to establish a secure and resilient battery supply chain by 2030. A significant focus of the blueprint is on large-scale end-of-life reuse and recycling of critical minerals to reinforce all stages of the domestic LIB supply chain [11]. Additionally, under the Core Exchange with Complete Vehicle Backstop policy, vehicle manufacturers are held accountable for ensuring that batteries from end-of-life EVs are either refurbished, reused, or recycled [12].

To improve traceability and transparency throughout the entire battery supply chain, both EU and China have initiated the development of battery passports. In 2022, the EU formally adopted the concept of a battery passport, a digital documentation system designed to store comprehensive information about each EV battery with a capacity exceeding 2 kWh. Cell manufacturers, module and battery producers, automotive OEMs, and companies involved in

battery servicing, refurbishing, and repurposing, contributed to the data. This initiative enables transparency regarding material sources and post-use status, thereby enhancing supply chain security and supporting the establishment of circular business models [13]. Following this lead, China launched its digital battery passport initiative in 2023 [14]. In parallel, the Global Battery Alliance (GBA) established a proof-of-concept framework in 2023, covering the entire value chain from mining to vehicle manufacturing. By 2024, GBA progressed to real-world data collection, engaging the world's 10 largest battery cell manufacturers to monitor material flows, gather and validate sustainability-related data, and develop Environmental, Social, and Governance (ESG) scoring methodologies [15].

Implementing a digital battery passport policy will strengthen supply chain traceability and transparency throughout the lithium-ion battery life cycle. By assigning each battery a unique coded digital identity that stores key data, such as the origin of raw materials, chemical composition, recycled content, and carbon footprint, the passport facilitates end-of-life logistics and compliance monitoring. Such a tracking system makes recycling enforcement easier and provides recyclers with the chemistry information needed to select optimal recovery routes and maximize metal yields.

Several jurisdictions, notably EU and China, have already mandated battery passports and set minimum recycling efficiency and recycled-content quotas for cobalt, nickel, and lithium, but wider international adoption remains necessary. Globally harmonized policies, particularly focusing on the inclusion of recycled materials in new cell production, would help reduce critical mineral supply risks, curb environmental impact, and accelerate EV deployment to meet GHG reduction goals.

2.4. Circular strategies: Reuse and Recycle

2.4.1. Reuse—Second Life applications

The reuse of LIBs, commonly referred to as second-life applications, offers a promising strategy for extending battery lifespan and supporting the circular economy. Reuse aims to extend the useful lifetime of batteries, lower the investment and operational costs of energy systems, and minimize the demand for raw materials [6]. After approximately 8–10 years of use in EVs, LIBs typically retain 80–85% of their original capacity, which is sufficient for secondary applications such as backup power systems. Once the capacity drops below 80% of its rated value, the battery is no longer considered suitable for EV use but can still serve in less demanding stationary energy storage roles [16,17]. In second-life applications, battery modules removed from EVs are repurposed and reconfigured into battery packs for a range of utility-oriented uses. These include stationary energy storage systems (SES) for grid-scale applications, building energy management, backup systems for telecommunications towers and data centers, and even as spare batteries for EVs [18]. According to Boston Consulting Group (BCG), by 2030, the global demand for batteries in SES is expected to reach 120 GWh annually, indicating significant potential for repurposed EV batteries. Moreover, studies estimate that second-life utilization can reduce CO₂ emissions by up to 65% compared to manufacturing new battery systems for similar applications [19].

Several commercial initiatives are exploring this pathway. For instance, Nissan, in collaboration with Ecobat Solutions UK Ltd., is investigating reuse options for batteries initially deployed in its LEAF model, first launched in 2011. The identified second-life applications include emergency power backup, grid power balancing, and mobile charging stations. These initiatives contribute to Nissan's broader target of achieving carbon neutrality by 2050 [20]. In Canada,

Moment Energy was awarded \$320,000 under the CleanBC Go Electric Advanced Research and Commercialization (ARC) Program to develop a low-carbon energy storage system utilizing repurposed EV batteries [21].

Despite its promise, battery reuse presents several challenges. The technical processes involved, such as dismantling, health assessment, disassembly, module clustering, and reassembly, can be resource-intensive and costly [19]. Profit margins are typically low; for instance, a case study on nickel-manganese-cobalt (NMC) and nickel-cobalt-aluminum (NCA) batteries reported an EBIT margin of less than 5%, which offers a limited financial incentive for large-scale industrial adoption [18]. Additionally, variability in battery design and architecture across manufacturers complicates automation in the disassembly process, further hindering scalability [19]. Due to these technical and economic barriers, some companies regard recycling as a more viable and efficient strategy for achieving circularity in the LIB lifecycle.

2.4.2. Recycle

Recycling refers to the collection and processing of waste materials into new, usable products [22]. In the context of batteries, this process involves recovering valuable metals from end-of-life EV batteries and converting them into precursor materials for new battery production. This approach reduces dependence on the extraction of critical minerals, which are fundamental to battery chemistry and directly influence energy density. According to IEA, scaling up battery recycling could decrease the demand for newly mined lithium and nickel by 25% and copper and cobalt by 40% by 2050. In addition to easing pressure on mineral reserves, recycling significantly lowers the environmental impacts associated with mining. For instance, the recycling of nickel, cobalt, and lithium results in up to 80% fewer GHG emissions compared to their primary extraction counterparts [23]. Furthermore, recycling helps mitigate geopolitical risks associated with the concentration of critical mineral reserves and refining operations in a few regions, thereby reducing the potential for mineral resource scarcity by approximately 25% [24].

The recycling of LIBs begins with pre-treatment, a critical step that involves the sorting of spent batteries based on their chemistry, form factor, density, and dimension [25]. The next step involves safe discharging to prevent thermal incidents, such as fires, and disassembling battery packs and modules to isolate individual cells and recover the black mass, a fine powder composed of active electrode materials.

The extracted black mass then undergoes metallurgical processing through one of three main approaches: pyrometallurgy, hydrometallurgy, or direct recycling.

- Pyrometallurgy involves the thermal treatment of black mass via roasting or smelting, converting the material into metal oxides or alloys [26]. This technique is well-established and has reached a high level of industrial maturity; companies such as Umicore have adopted it at a commercial scale. However, pyrometallurgy inefficiently recovers specific components, such as lithium, electrolytes, separators, and graphite, which are often lost during the high-temperature process [27].
- Hydrometallurgy relies on chemical leaching, using inorganic acids (e.g., H_2SO_4 , HCl , HNO_3) or organic acids (e.g., citric and oxalic acids) to dissolve valuable metals into solution. These metals are subsequently precipitated and purified [26]. This method offers several advantages: high recovery efficiency, low impurity levels, and cost-effectiveness, making it the most widely adopted recycling process in the battery industry today [27].

- Direct recycling aims to restore the performance of degraded electrode materials through non-destructive rejuvenation techniques without breaking them down into elemental constituents. Standard methods include solid-state relithiation, electrochemical relithiation, ionic liquid-assisted relithiation, eutectic salt relithiation, and solution-based relithiation [28]. This approach is considered environmentally superior due to its minimal energy consumption and reduced reliance on corrosive chemicals. While individual recycling operations may employ one or a combination of these methods, industrial uptake of direct recycling remains limited due to scalability challenges.

An LCA of NMC 811 battery recycling using various methods showed that direct recycling yields the highest reduction in GHG emissions (-50.94 kg CO₂-eq), followed by hydrometallurgy (-33.12 kg CO₂-eq), and finally pyrometallurgy, which offered the least environmental benefit (-9.46 kg CO₂-eq) [29]. These findings underscore the significant environmental potential of direct recycling; however, due to current technical and logistical constraints, its widespread industrial application remains limited. Consequently, hydrometallurgy stands out as a promising alternative, striking a balance between environmental impact and recycling efficiency.

The growing recognition of the benefits of battery recycling has prompted many nations to implement supportive policies and regulatory frameworks. Among the global leaders in this domain are the EU and China, both of which have adopted ambitious strategies to promote circularity in battery value chains. The EU has mandated that, by 2031, all LIBs with capacities exceeding 2 kWh must contain minimum recycled content thresholds: 16% for cobalt, 6% for lithium, and 6% for nickel. In addition, the EU has set progressive recycling efficiency targets for 2030: $\geq 90\%$ for cobalt, copper, and nickel, and $\geq 50\%$ for lithium [30]. China also plays a dominant role in global battery recycling infrastructure, is projected to account for 78% of battery pre-treatment capacity and 89% of black mass refining capacity by 2025, translating to the processing of approximately 895 kilotons of black mass [31]. Recycled materials will fulfill 13% to 18% of China's total battery metal demand in 2025 [32]. Moreover, China has established stringent recovery rate targets: at least 90% for lithium and no less than 98% for nickel, cobalt, and manganese [33]. Reinforcing its commitment to technological advancement, the country mandates that recycling enterprises allocate a minimum of 3% of their annual revenue toward R&D and process improvement initiatives [34]. These concerted efforts highlight the strategic importance of battery recycling, not only as a means of resource conservation but also as a critical pillar in achieving a sustainable, circular economy in the battery sector.

Despite its advantages, recycling is not without challenges. Advancements in recycling technologies are crucial for enhancing the recovery rates of battery metals. Achieving high recovery efficiency is important not only from an economic standpoint but also to minimize the need for virgin material inputs. Additionally, the diversity in cell designs and manufacturing processes presents obstacles to efficient recycling. Enhancing automation technologies could help alleviate these limitations [35]. Recycling remains a key component in enabling the circular economy, and overcoming these existing challenges is critical to realizing closed-loop LIB production and broader adoption of circular practices.

2.5. Concluding remarks and perspectives

The growing global population is placing increasing strain on natural resources, resulting in higher material consumption, greater product manufacturing, and elevated waste generation. In this context, implementing circular economy principles is essential for achieving long-term

environmental sustainability, particularly for resource-intensive technologies such as LIBs. Among the various circular strategies, recycling is the most widely adopted, primarily due to its economic advantages. However, recycling critical battery minerals presents several challenges, necessitating ongoing research and innovation to achieve the high recycling efficiency targets set by certain countries. Moreover, recycling processes must become more environmentally sustainable to align with the broader objective of making the entire LIB value chain both ecologically and economically viable.

While the EU and China have taken commendable steps by implementing policies that support circularity in LIBs, there remains a pressing need for globally harmonized regulations that ensure a closed-loop supply chain. Mandating minimum recycled content in new cell production can significantly alleviate pressure on critical mineral supply while also reducing environmental impacts. Additionally, initiatives such as the battery passport, which documents both the origin of raw materials and ESG data, offer valuable transparency. Global enforcement of such tools would greatly enhance traceability, accountability, and sustainability across the LIB lifecycle.

References

1. “What is a circular economy? | Ellen MacArthur Foundation.” Accessed: Jun. 18, 2025. [Online]. Available: <https://www.ellenmacarthurfoundation.org/topics/circular-economy-introduction/overview>
2. “Circular economy: definition, importance and benefits | Topics | European Parliament.” Accessed: Jun. 18, 2025. [Online]. Available: <https://www.europarl.europa.eu/topics/en/article/20151201STO05603/circular-economy-definition-importance-and-benefits>
3. “Circular economy action plan - European Commission.” Accessed: Jun. 18, 2025. [Online]. Available: https://environment.ec.europa.eu/strategy/circular-economy-action-plan_en
4. R. Chen, S. Vlieger De Oliveira, and C. Mahut, “China’s Circular Economy Policies: Review and Reflection A CIRCULAR ECONOMY VISION,” 2023.
5. J. N. Meegoda, S. Malladi, and I. C. Zayas, “End-of-Life Management of Electric Vehicle Lithium-Ion Batteries in the United States,” *Clean Technologies* 2022, Vol. 4, Pages 1162–1174, vol. 4, no. 4, pp. 1162–1174, Nov. 2022, doi: 10.3390/CLEANTECHNOL4040071.
6. S. A. Jose et al., “Pathways to Circular Economy for Electric Vehicle Batteries,” *Recycling* 2024, Vol. 9, Page 76, vol. 9, no. 5, p. 76, Sep. 2024, doi: 10.3390/RECYCLING9050076.
7. M. Goyal, K. Singh, and N. Bhatnagar, “Circular economy conceptualization for lithium-ion batteries- material procurement and disposal process,” *Chem Eng Sci*, vol. 281, p. 119080, Nov. 2023, doi: 10.1016/J.CES.2023.119080.
8. Connor and Phoebe, “An Analysis of Lithium-ion Battery Fires in Waste Management and Recycling,” 2021.
9. “Pathways To Achieve New Circular Vision for Lithium-Ion Batteries | NREL.” Accessed: Jun. 20, 2025. [Online]. Available: <https://www.nrel.gov/news/detail/program/2021/pathways-to-achieve-new-circular-vision-for-lithium-ion-batteries>
10. “Battery-related waste codes update set to boost circular economy - European Commission.” Accessed: Jun. 30, 2025. [Online]. Available: https://environment.ec.europa.eu/news/battery-related-waste-codes-update-set-boost-circular-economy-2025-03-05_en?utm_source=chatgpt.com
11. “National Blueprint for Lithium Batteries | Department of Energy.” Accessed: Jun. 30, 2025. [Online]. Available: <https://www.energy.gov/eere/vehicles/articles/national-blueprint-lithium-batteries>
12. “EXECUTIVE SUMMARY”, Accessed: Jun. 30, 2025. [Online]. Available: <https://www.autosinnovate.org/posts/papers-reports/Get%20Connected%20EV%20Quarterly%20Report%20Q4.pdf>
13. “EU Sustainable Batteries Regulation – Policies - IEA.” Accessed: Jun. 30, 2025. [Online]. Available: <https://www.iea.org/policies/16763-eu-sustainable-batteries-regulation>
14. “Digital Battery Passports: An Enabler for Sustainable and Circular Battery Management”.
15. “Battery Passport.” Accessed: Jun. 30, 2025. [Online]. Available: <https://www.globalbattery.org/battery-passport/>
16. “A circular economy for batteries to underpin renewable energy growth.” Accessed: Jun. 21, 2025. [Online]. Available: <https://www.ellenmacarthurfoundation.org/articles/a-circular-economy-for-batteries-to-underpin-renewable-energy-growth>
17. M. Pagliaro and F. Meneguzzo, “Lithium battery reusing and recycling: A circular economy insight,” *Heliyon*, vol. 5, no. 6, p. e01866, Jun. 2019, doi: 10.1016/j.heliyon.2019.e01866.

18. "The Case for a Circular Economy in Electric Vehicle Batteries | BCG." Accessed: Jun. 22, 2025. [Online]. Available: <https://www.bcg.com/publications/2020/case-for-circular-economy-in-electric-vehicle-batteries>
19. M. K. Al-Alawi, J. Cugley, and H. Hassanin, "Techno-economic feasibility of retired electric-vehicle batteries repurpose/reuse in second-life applications: A systematic review," *Energy and Climate Change*, vol. 3, p. 100086, Dec. 2022, doi: 10.1016/J.EGYCC.2022.100086.
20. "Nissan and Ecobat to give used EV batteries a second life beyond the car." Accessed: Jun. 21, 2025. [Online]. Available: <https://uk.nissannews.com/en-GB/releases/nissan-and-ecobat-to-give-used-ev-batteries-a-second-life-beyond-the-car>
21. "BC Gov News." Accessed: Jun. 21, 2025. [Online]. Available: <https://news.gov.bc.ca/releases/2022EMLI0020-000512>
22. "Recycling Basics and Benefits | US EPA." Accessed: Jun. 23, 2025. [Online]. Available: <https://www.epa.gov/recycle/recycling-basics-and-benefits>
23. "Executive summary – Recycling of Critical Minerals – Analysis - IEA." Accessed: Jun. 22, 2025. [Online]. Available: <https://www.iea.org/reports/recycling-of-critical-minerals/executive-summary>
24. M. Rezaei et al., "A review of lithium-ion battery recycling for enabling a circular economy," *J Power Sources*, vol. 630, p. 236157, Feb. 2025, doi: 10.1016/J.JPOWSOUR.2024.236157.
25. R. P. Sheth, N. S. Ranawat, A. Chakraborty, R. P. Mishra, and M. Khandelwal, "The Lithium-Ion Battery Recycling Process from a Circular Economy Perspective—A Review and Future Directions," *Energies* 2023, Vol. 16, Page 3228, vol. 16, no. 7, p. 3228, Apr. 2023, doi: 10.3390/EN16073228.
26. Z. J. Baum, R. E. Bird, X. Yu, and J. Ma, "Lithium-Ion Battery Recycling—Overview of Techniques and Trends," *ACS Energy Lett*, vol. 7, no. 2, pp. 712–719, Feb. 2022, doi: 10.1021/ACSENERGYLETT.1C02602/ASSET/IMAGES/LARGE/NZ1C02602_0005.JPEG.
27. Y. Wang et al., "Recent progress on the recycling technology of Li-ion batteries," *Journal of Energy Chemistry*, vol. 55, pp. 391–419, Apr. 2021, doi: 10.1016/J.JECHEM.2020.05.008.
28. P. Xu, D. H. S. Tan, B. Jiao, H. Gao, X. Yu, and Z. Chen, "A Materials Perspective on Direct Recycling of Lithium-Ion Batteries: Principles, Challenges and Opportunities," *Adv Funct Mater*, vol. 33, no. 14, p. 2213168, Apr. 2023, doi: 10.1002/ADFM.202213168;REQUESTEDJOURNAL:JOURNAL:16163028;WGROU: STRING:PUBLICATION.
29. Q. Chen, X. Lai, H. Gu, X. Tang, F. Gao, X. Han, and Y. Zheng, "Investigating carbon footprint and carbon reduction potential using a cradle-to-cradle LCA approach on lithium-ion batteries for electric vehicles in China," *Journal of Cleaner Production*, vol. 369, p. 133342, Oct. 2022.
30. "Regulation - 2023/1542 - EN - EUR-Lex." Accessed: Jun. 24, 2025. [Online]. Available: <https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32023R1542>
31. "Infographic: How much of the battery recycling industry does China control? | Benchmark Source." Accessed: Jun. 24, 2025. [Online]. Available: <https://source.benchmarkminerals.com/article/infographic-how-much-of-the-recycling-industry-does-china-control>
32. W. Pan, "Chinese Policy activities with respect to battery sustainability and battery ID," 2024.

33. “China unveils new used EV power batteries rules; recycling market remains gloomy | S&P Global.” Accessed: Jun. 24, 2025. [Online]. Available: <https://www.spglobal.com/commodity-insights/en/news-research/latest-news/metals/081624-china-unveils-new-used-ev-power-batteries-rules-recycling-market-remains-gloomy>
34. “Specifications for the Comprehensive Utilisation of Waste EV Batteries 2024 – Policies - IEA.” Accessed: Jun. 24, 2025. [Online]. Available: <https://www.iea.org/policies/24987-specifications-for-the-comprehensive-utilisation-of-waste-ev-batteries-2024>
35. X. Yu et al., “Current Challenges in Efficient Lithium-Ion Batteries’ Recycling: A Perspective,” *Global Challenges*, vol. 6, no. 12, p. 2200099, Dec. 2022, doi: 10.1002/GCH2.202200099;WGROU:STRING:PUBLICATION.

Chapter 3 - Transformation from ore to battery-grade salts

This chapter provides a comprehensive review of the critical minerals essential for clean energy transitions, with a specific focus on nickel and lithium. It explores the pathway from nickel extraction to the conversion of both nickel and lithium into battery-grade salts, with a particular emphasis on environmentally sustainable metallurgical processing technologies.

3.1. Critical Minerals

Critical minerals are the minerals identified as essential by individual government bodies or organizations due to their strategic importance in development, particularly for emerging technologies and the transition to green energy systems. The classification of a mineral as "critical" is not standardized and depends on multiple factors, including geological availability, economic importance, technological applicability, availability of substitutes, environmental impacts, and end-use applications. Over time, researchers have proposed quantitative frameworks to assess mineral criticality, typically based on three core dimensions: (1) the impact of supply constraints, (2) associated environmental risks, and (3) the likelihood of supply disruptions [1]. According to the IEA, there are 37 critical minerals vital to achieving clean energy transitions. In the context of LIBs, the key critical minerals are lithium, nickel, cobalt, manganese, and graphite [2].

The strategic importance of these minerals necessitates not only their efficient recycling but also the adoption of sustainable mining practices. In recognition of this need, the Sustainable Critical Minerals Alliance was established during COP15 in Montreal to promote environmentally responsible mining and support circular economy approaches for critical minerals [3]. According to IEA projections, meeting the NZE target by 2050 will require approximately 2364.2 kilotons of nickel and 1446.8 kilotons of lithium for EV applications alone [2]. The 2040 forecast indicates a significant gap between projected demand and available supply. Consequently, the deployment of zero-waste mining technologies, in conjunction with large-scale recycling, is crucial to bridging this gap and ensuring long-term sustainability.

3.2. Mineral resources and reserves

3.2.1. Lithium

The unique properties of LIBs have changed the field of energy storage technology; consequently, the demand for lithium has increased over the years. According to the latest reports from the U.S. Geological Survey, the total lithium resources on Earth are approximately 105 million tons, of which 28 million tons are present in lithium reserves. The renowned "lithium triangle," comprising Bolivia, Chile, and Argentina, holds the most significant lithium resources (brine deposits), accounting for 56 million tons. Next, the United States (brine and clay deposits) with 14 million tons, Australia (pegmatite ore deposits) with 8.7 million tons, and China (brine deposits) with 6.8 million tons [4].

Lithium is extracted from three types of deposits: (1) pegmatite deposits, (2) continental brines and oilfields, and (3) hydrothermally altered clays. The grade or quality of the lithium obtained from the deposits is the highest in pegmatite deposits and the lowest in brines. Pegmatites have several forms, such as spodumene, petalite, zinnwaldite, and lepidolite; out of these forms, spodumene (contains 6–9 wt% Li_2O) is the most economical form for obtaining the battery-grade material for lithium-rich CAM in the form of lithium hydroxide or lithium carbonate [5]. The ubiquitous availability of pegmatite deposits results in less susceptibility to supply chain disruptions. However, for long-term production, there is a need for extensive exploration, which might become costlier [6]. Lithium clay deposits are further divided into (1) hectorite, (2) jadarite, and (3) ion-clay deposits. Brine solutions, such as hypersaline water, are typically the preferred source of lithium because they have the potential for large-scale and long-term production compared with pegmatite deposits [5]. Several factors, such as recovery from brine without dilution and degradation of reservoir rocks, could restrict the extraction capacity of lithium from brines [6]. From the perspective of cost and margin, brine extraction has a margin of USD 5386/t LCE, which is nearly double that of pegmatite operations [7]. Additional research advancements in brine extraction could enhance its potential and aid in meeting the increasing demand for lithium.

Within the lithium triangle, Bolivia possesses the largest brine deposits; however, political and financial challenges in Bolivia have hindered the full exploitation of its "white gold" potential [8]. China has 80% of the world's lithium refineries, which poses challenges for a sustainable lithium supply chain. Transporting battery-grade lithium from China to North America and Europe also contributes to GHG. Additionally, escalating geopolitical tensions between China and Western nations may lead to export restrictions on lithium, as already seen with natural graphite [9]. The geopolitical tensions have given rise to developing a local lithium supply chain in North America. In the Lithium Triangle, water levels in the salars are critically low, and with 400,000 L of water needed to produce 1 ton of lithium, this extraction process further escalates this issue. Moreover, lithium mining effluents are degrading water quality and harming local biodiversity [10].

3.2.2. Nickel

The primary utilization of nickel is in the form of stainless steel. However, battery manufacturers use nickel in nickel–cadmium and nickel–metal hydride batteries, and interest in LIBs as NMC and NCA chemistries is growing. Nickel provides high energy density and high storage capacity at low cost; consequently, the CAM chemistry is moving toward 90% nickel content [11].

Mining operations extract nickel from two types of ores: laterite and sulfide ores. Laterite ores are more complex to process than sulfide ores such as pentlandite; hence, there has been a predefined inclination toward sulfide ores, but the growing demand for nickel in batteries has increased the focus on laterite ores. Sulfide ores are formed from volcanic or hydrothermal processes, whereas laterite ores are formed by the weathering of ultramafic rocks [12]. The moisture content of laterite makes it challenging to process, whereas the sulfur content of sulfide ores reduces the energy requirements [13]. Laterite ores can be categorized as limonite (lower nickel (1.1%), high iron, and low cobalt content) and saprolite (higher nickel (2%), high magnesium, and low iron) [14]. Laterite ore mining requires less energy; however, its processing consumes a significant amount of energy. Nickel also occurs in polymetallic nodules on the ocean floor. In total, there are 350 million tons of nickel resources available globally, out of which 54% are laterite ores and 35% are sulfide ores. More than 130 million ore reserves are available for nickel extraction, with Indonesia (laterite ores) accounting for 55 million tons, followed by Australia (both sulfide and laterite ores) and Brazil (laterite ores) at 24 million tons and 16 million tons, respectively [4]. A growing number of battery recycling industries can contribute to the availability of Ni in the form of battery-grade nickel to meet the demand for batteries [15].

The fluctuating price of nickel is significantly affected the laterite ore mining operations. For instance, Indonesian mining companies recently demonstrated the conversion of low-grade class II nickel into high-purity LME-grade cathodes, which has driven the price of mixed hydroxide precipitate (MHP) below \$15,000 per metric ton [16]. HPAL achieved this advancement; however, its tailings have a significant environmental impact in Indonesia [17]. Apart from technological advancement, geopolitical tensions also impact nickel prices. The ongoing conflict between Russia and Ukraine has further worsened the price fluctuations [18]. Sulfide ores have the potential to produce valuable byproducts, such as Co, Cu, and Pt, which give them an edge over laterite ore. Hence, sulfide ore is more profitable than laterite ore. As of 2021, the realized Ni mining price was approximately USD 8/lb [19]. New technological advancements can affect nickel prices and, consequently, dominate mining operations.

3.3. Transformation from ore to battery-grade salts

The production of battery-grade salts generally involves three key stages: beneficiation, metallurgical processing, and purification or refining, as illustrated in Fig. 1. This section provides a concise overview of the transformation processes employed for various nickel and lithium sources.

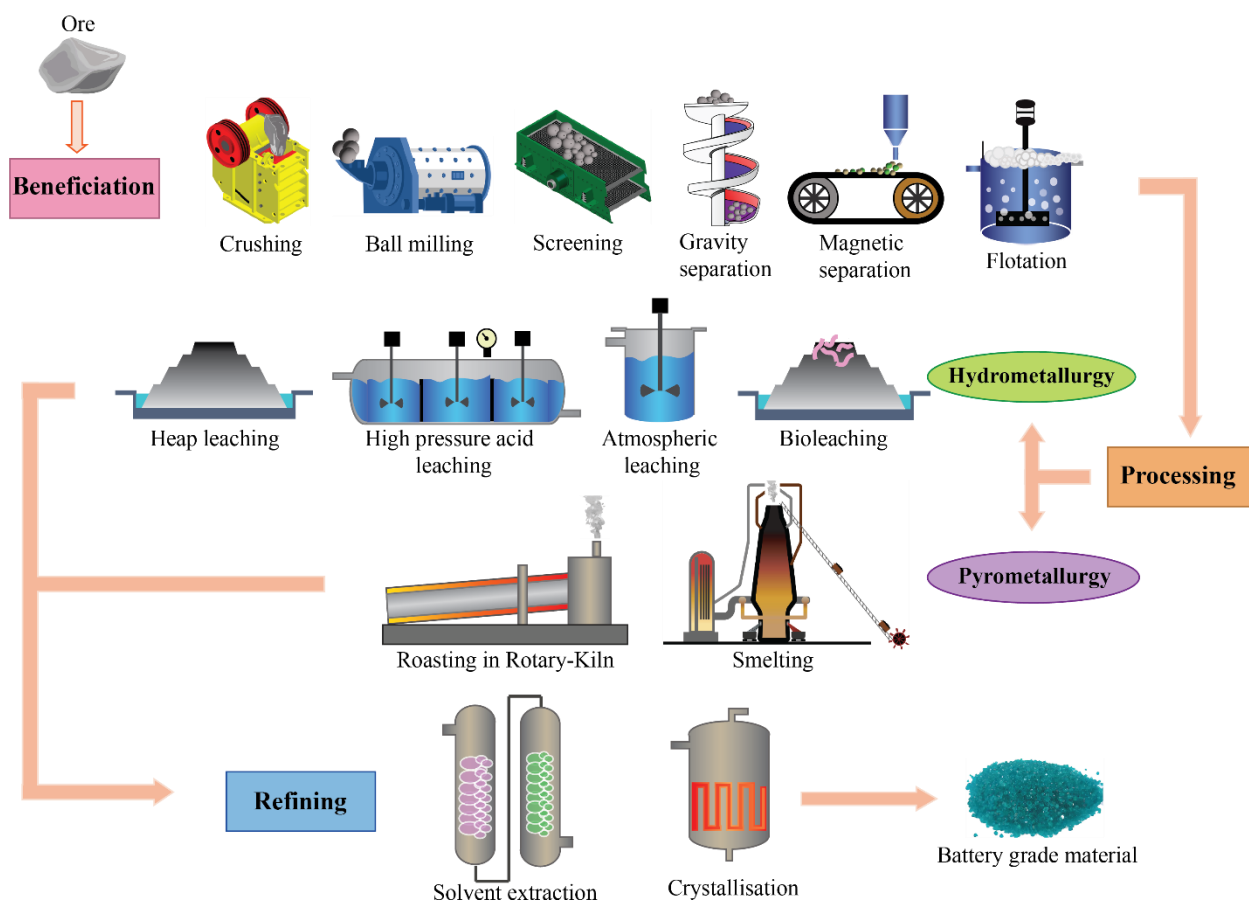


Figure 1: Overview of the general process and methods used for extracting battery-grade materials from ores. The process consists of multiple steps, from beneficiation to processing by either hydrometallurgical or pyrometallurgical methods depending on the type of ore to refining of the intermediate product to produce the battery grade material.

3.3.1. Lithium sources

During cathode active material production, lithium is typically added during calcination to provide Li ions to the LIBs. Commonly used battery-grade lithium salts for LIB synthesis include lithium carbonate (Li_2CO_3) and lithium hydroxide monohydrate ($\text{LiOH}\cdot\text{H}_2\text{O}$). For nickel-rich cathodes, Li_2CO_3 requires calcination at a high temperature (850–900°C), which can compromise its crystal structure. Consequently, there is a growing preference for $\text{LiOH}\cdot\text{H}_2\text{O}$, which allows calcination at a lower temperature range (650–700 °C). Producers further process Li_2CO_3 to produce $\text{LiOH}\cdot\text{H}_2\text{O}$ through electrochemical, pyrometallurgical, ion-exchange, or hydrometallurgical methods [20]. Spodumene ore and brine are the primary sources of lithium, with each source undergoing a different process to obtain battery-grade lithium salts, as shown in Fig. 2. In this section, we briefly outline the processing steps for ore and brine.



Figure 2: General overview of extraction process of battery-grade materials from lithium from spodumene and brine

3.3.1.1. Spodumene ores

The processing of spodumene ore begins with crushing and grinding the ore using jaw crushers and ball milling to liberate the minerals by reducing the particle size. The next step is flotation, which increases the concentration of Li_2O from 1% to 4%–6%, achieved using dense nonaqueous liquids. The concentrate can be further purified through other beneficiation methods, such as dense media separation, magnetic separation, and gravity concentration, to remove impurities and obtain α -spodumene [21]. In the traditional sulfuric acid method, heating α -spodumene in a rotary kiln at $1,100^\circ\text{C}$ converts it to β -spodumene. Treating β -spodumene with 30%–40% excess H_2SO_4 at 200 – 250°C produces lithium sulfate, which is then reacted with sodium carbonate to yield Li_2CO_3 [22]. The traditional processing of spodumene ore has a GWP ranging from 17.11 to 22.33 kg CO_2 -eq per kg of Li_2CO_3 , with water consumption between 0.2 and 0.5 m^3 per kg of Li_2CO_3 for an Australian mine and refining facility in China [23]. The process of converting α -spodumene to β -spodumene is known as calcination and is employed because α -spodumene is resistant to chemical reactions.

β -spodumene can be processed through multiple pathways to produce battery-grade lithium salts, including roasting followed by leaching or direct leaching. Various roasting techniques, such as sulfation, carbonation, and chlorination, employ different additives, including sulfate, carbonate, and chlorine-based salts. The choice of roasting method directly influences the type of lithium compound obtained, resulting in products such as lithium sulfate (Li_2SO_4), lithium carbonate (Li_2CO_3), lithium hydroxide (LiOH), or lithium chloride (LiCl) [21]. Direct leaching of β -spodumene is another method that has also been explored, such as leaching of β -spodumene using HF , which achieved an extraction efficiency of 90%. However, the Li_2CO_3 crystals obtained had a purity of only 98.3%, which is insufficient for battery-grade applications [24]. As the calcination process requires a high amount of energy, direct transformation of α -spodumene to Li_2CO_3 has gained attention. Wang S treated α -spodumene directly with Na_2CO_3 and Al_2O_3 at 750°C for 4 h, achieving a 90% recovery of Li_2CO_3 . The addition of Al_2O_3 was crucial for preventing the excessive formation of Li_2SiO_3 , which would occur if only Na_2CO_3 were used [25].

In the pursuit of more environmentally sustainable solutions, electrochemical leaching has emerged as an effective alternative processing method. In their study, α -spodumene or β -spodumene was mixed with conductive carbon and a PVDF binder and then coated onto a graphite rod. In addition, a 3D current collector modified with a gold catalyst was developed. Electrochemical leaching in 0.5 M H_2SO_4 employed H_2O_2 as a promoter to lower the reaction potential. This method achieved a leaching efficiency of 92.2% and reduced CO_2 emissions by 75.3% compared to traditional processes [26]. This study highlights the significant potential of electrochemical leaching over conventional leaching methods.

3.3.1.2. Brine

Brine typically contains low lithium concentrations ranging from 0.3% to 1.5%. To extract lithium, operators pump brine into evaporation ponds, removing 90% of the water using solar or wind evaporation. This process is time-intensive and often takes up to a year to reach the desired lithium concentration of 6%–7%. Upon reaching this concentration, Na_2CO_3 precipitates the lithium carbonate, which converts into lithium hydroxide after processing. In regions such as the Lithium Triangle, the GWP of this processing method can range from 5 to 25 kg CO_2 -eq per kg of Li_2CO_3 , depending on the ore grade. Water consumption varies between 0.2 and 7.7 m^3 per kg of Li_2CO_3 [23]. To address the environmental and temporal limitations of conventional brine extraction, several direct lithium extraction (DLE) technologies have been investigated [27]. These techniques selectively extract lithium from brine using methods such as adsorption, ion exchange, solvent extraction, membrane separation, and electrochemical processes.

- Adsorption - This method involves the physical or chemical binding of lithium ions to the surface of solid adsorbents [28]. The adsorbed lithium is then desorbed using either chemical reagents or water [29]. Effective adsorbents must exhibit high lithium selectivity, adequate adsorption capacity, strong operational stability, and economic feasibility. For adsorption-based DLE to be viable, brine must contain lithium concentrations exceeding 100 mg/L, along with sufficient salinity and elevated temperatures. Aluminium-based adsorbents have shown 80–90% lithium extraction efficiency across various brine compositions, demonstrating their broad applicability [30].
- Ion exchange - In this approach, lithium ions are exchanged with similarly charged cations from specially designed resins. Acid then recovers the lithium [28]. These resins contain functional groups that enable preferential lithium uptake. Resins based on aluminium, magnesium, and titanium have been explored for this purpose [28, 31].
- Solvent extraction - Mixing brine with an organic solvent extractant selectively extracts lithium by dissolving Li^+ ions while rejecting others. Acid then strips the lithium-enriched extractant to recover lithium [32]. Common extractants include kerosene, m-xylene, p-xylene, chlorobenzene, benzene, dodecane, chloroform, cyclohexane, and various ethers. Extraction efficiency depends on pH, temperature, contact time, solvent type, and number of extraction cycles [28]. For example, the TBP– FeCl_3 extractant, when tested on Chilean brine, achieved 74.2% lithium extraction after four stages and 80.3% recovery after ten scrubbing stages, indicating high efficacy [33].
- Membrane separation - This technique employs pressure-driven membrane processes such as ultrafiltration (UF), microfiltration (MF), or nanofiltration (NF), the latter being most effective for lithium extraction. NF operates via size exclusion and charge-based repulsion (Donnan exclusion), effectively rejecting multivalent ions such as Mg^{2+} while allowing

lithium to pass through [33]. Li et al. enhanced lithium–magnesium separation by incorporating 15-crown-5 ether into a polyamide NF membrane, achieving a separation factor ($S_{\text{li,mg}}$) of 14 for a brine with a $\text{Mg}^{2+}/\text{Li}^{+}$ mass ratio of 20 [34]. Due to its high rejection of multivalent ions, NF also reduces membrane fouling and allows partial water recovery [33].

- Electrochemical methods - This method includes electrodialysis and capacitive deionization (CDI). In electrodialysis, cation-exchange membranes separate lithium, while in CDI, electrodes selectively adsorb lithium and later desorb it. Key parameters for performance evaluation include lithium selectivity coefficient, separation factor, salt adsorption capacity, and specific energy consumption [35]. In one study, Torres et al. designed a three-compartment electrodialysis reactor comprising a titanium mesh anode coated with iridium-based metal oxide, a stainless-steel mesh cathode, and a central brine chamber to obtain Li_2CO_3 with a purity of 93.8%–97.5% [36].

DLE methods have demonstrated significantly lower environmental impacts compared to traditional brine evaporation. An LCA of various DLE technologies, including solvent extraction, adsorption, NF, and membrane electrolysis, identified NF as the most environmentally sustainable, with a GWP of 17.7 kg CO_2 -eq per kg of Li_2CO_3 [37]. Owing to its environmental benefits and operational performance, NF presents a promising route for making lithium extraction from brines more sustainable.

3.3.2. Nickel sources

High-purity Class I nickel (>99.8% Ni), which includes forms such as briquettes (manufactured by sintering and pressing nickel powder), powders, cathodes, and oxides, produces battery-grade nickel sulfate. Additionally, nickel intermediates with lower nickel content (35%–70%), such as MHP, mixed sulfide precipitate (MSP), and nickel matte, can also produce battery-grade nickel sulfate [38]. The two types of nickel ores, sulfide and laterite, undergo different processes, as demonstrated in Fig. 3. For sulfide ores, beneficiation usually involves flotation. In contrast, laterite ores are treated directly through hydrometallurgical routes.



Figure 3: General overview of extraction process of battery-grade materials from nickel from laterite and sulfide ores

3.3.2.1. Sulfide ores

Associated minerals, such as Cu–Co–PGE and gangue minerals, influence the processing of sulfide ores. Typically, the process begins with beneficiation, in which various methods, including crushing, multistage grinding, gravity separation, and flotation (Fig. 3), are employed, depending on the mineral content [39]. Fine grinding is occasionally employed to liberate secondary minerals [40]. Flotation is an important step in improving the grade of Ni in sulfide ores. In this process, frothers and collectors are added to the flotation unit, along with a continuous supply of air to create bubbles. The sulfide minerals rise with the frother and are collected at the top, whereas the gangue minerals settle at the bottom. In copper-nickel sulfide ores, the presence of hydrophilic serpentine complicates Cu-Ni/MgO separation, which further affects the smelting efficiency. To address these issues, researchers have investigated the use of Sodium hexametaphosphate. Zhao K et al. demonstrated that sodium phytate improved Ni recovery from 6.86% Ni at 69.01% recovery to 8.03% Ni at 71.44% recovery from copper-sulfide ore, while reducing MgO content from 9.07% to 5.78% [41].

After flotation, electric furnaces or flash smelters smelt the Ni concentrate to produce Ni matte. Smelting in an oxidized atmosphere converts iron to iron oxide, which reacts with silica to form an immiscible iron-silica slag that is removed separately [42]. Every ton of nickel produced generates approximately 6–16 tons of slag and 4 metric tons of SO₂. [43, 44]. Canada mines sulfide ores and processes them through the pyrometallurgical route to produce matte, which Norway further refines, resulting in a total emissions impact of 6.72 kg CO₂ eq per kg NiSO₄·6H₂O. Among the various stages, matte production from concentrate has the highest contribution, accounting for 5.4 kg CO₂ eq [45]. Researchers have explored alternative methods, such as retaining sulfur in the solid state, to reduce SO₂ emissions. F. Wang et al. investigated mixing magnetite and activated carbon with nickel concentrate to reduce SO₂ emissions [42]. Converters further refine low-nickel

matte to remove impurities and produce high-nickel matte, which hydrometallurgical treatment then converts into battery-grade nickel. Various leaching techniques, such as nonoxidative H_2SO_4 leaching, chloride leaching, atmospheric oxidative leaching, and SO_2 reductive acid leaching, have been studied to obtain battery-grade nickel sulfate [46, 47, 48]. The impurities present in the leached solution are removed by employing different strategies such as the precipitation method, solvent extraction, and ion resin exchange [49, 50]. The precipitation method removes Fe, Al, and Cr by adjusting the pH of the solution. Hence, solvent extraction or ion resin exchange techniques can serve as alternatives to the precipitation method, as precipitation of Mg can lead to nickel losses.

The high demand for nickel and the depletion of high-grade sulfide ores have prompted research into the processing of low-grade sulfide ores. The processing of low-grade ores is challenging due to the presence of MgO silicate gangue, which complicates beneficiation, and a significant amount of Co is lost during smelting [51]. A combination of pyrometallurgical and hydrometallurgical methods, such as roasting and leaching, is often employed. Like the smelting process, the roasting process generates SO_2 , which requires the addition of sulfur-fixing agents like ZnO, CaO, or MgCO_3 . Sulfur and chlorination roasting are common techniques. Sulfur roasting offers advantages such as lower temperature, reduced acid consumption, and simpler operation [52]. Mu W et al. studied the effect of using MgCO_3 as a sulfur-fixing agent and achieved 98.5% nickel and 99% copper recovery after leaching with sulfuric acid [53]. Chlorinated roasting offers the advantages of high reaction efficiency. Xu X et al. achieved 83.6% Ni and 81.3% Cu recovery using reagents like NaCl and MgCl_2 for chlorination roasting, followed by water leaching [54].

Meng developed a compact process for producing NMC 811 CAM directly from nickel-copper sulfide ore. The process involved (1) ammonia leaching in the presence of oxygen, (2) boiling the lixivium, (3) removing Cu by MnS at 60°C , (4) precipitating nickel hydroxide by adding NaOH, and (5) synthesizing NMC 811 by the coprecipitation method. The material exhibited superior cycling performance (151 mAh g^{-1} with capacity retention of 84.2%) after 200 cycles compared with conventional samples (148 mAh g^{-1} capacity with capacity retention of 82.1%) [55]. Direct production of NCM CAM from sulfide ore could streamline the supply chain for battery-grade nickel by eliminating several intermediate processing steps, ultimately reducing the environmental impact observed due to transportation. Kinnunen et al. modeled a direct hydrometallurgical process for producing battery-grade nickel sulfate by incorporating chemical leaching, impurity removal by precipitation, solvent extraction, and crystallization. They also conducted an LCA analysis of the process. They found that the process showed increased chemical consumption and waste generation due to the iron content, thereby contributing to higher CO_2 emissions. However, it also offers the benefit of Cu recovery [56]. Optimizing and reducing reagent use could classify direct leaching of sulfide ore as a greener extraction process.

3.3.2.2. *Laterite ores*

Low Ni concentrations and isomorphic crystalline structures, such as goethite or serpentine, in laterite ores create challenges for flotation; hence, these ores directly undergo hydrometallurgical or pyrometallurgical processing [39]. However, researchers propose that fine grinding of laterite ores liberates minerals and facilitates flotation in cells, such as the reactor-separator, which provides superior particle–bubble attachment [57]. Testing these flotation cells can provide valuable insights into the flotation capabilities of laterite ores. Another technique for facilitating the beneficiation process is the conversion of laterite ore to sulfide ore. Sulfidation-flotation technology involves the introduction of sulfide agents, such as pyrite and sulfur, to the ore, followed by sulfidation roasting to convert the oxide ore into a sulfide ore [58]. As previously

mentioned, researchers classify laterite ores as saprolite and limonite, each requiring a different processing method. Pyrometallurgical processes are commonly used for saprolite ores, whereas hydrometallurgy or a combination of both pyrometallurgy and hydrometallurgy processes limonite ores [59]. The grade of the ore also influences the processing route; high-grade ores (>2% Ni) are processed pyrometallurgically, whereas low-grade ores (<1.3% Ni) are treated hydrometallurgically [60].

Common hydrometallurgical methods for processing laterite ores include heap leaching, atmospheric leaching, high-pressure acid leaching (HPAL), direct Ni processing, and bioleaching [61]. Researchers most widely use sulfuric acid as a leaching agent, and they have also investigated nitric acid, hydrochloric acid, and specific organic acids. Heap leaching is a simple and low-cost method in which ore is dumped into a heap and treated with acid, typically in an open atmosphere [62]. Atmospheric leaching occurs at ambient temperature and pressure in open vessels, eliminating the need for autoclaving [63]. Although the HPAL method is more expensive because of the use of an autoclave, it has the advantage of precipitating iron and aluminum as hematite and alunite, thereby regenerating acid [64]. The Direct Nickel process, developed by Direct Nickel Ltd., involves atmospheric leaching of laterite ore with nitric acid [65]. Bioleaching, which has attracted significant interest in recent years, involves the use of microorganisms to leach metals [66].

3.3.2.2.1. Limonite ores

Hydrometallurgical methods are generally applied to process limonite ore; however, depending on the mineralogical composition, pyrometallurgical methods may also be suitable. Pyrometallurgical techniques are preferred when the SiO₂/MgO ratio is between 10 and 12, whereas hydrometallurgical methods are preferred at lower ratios [67]. The previously mentioned hydrometallurgical methods commonly process limonite ore, with HPAL being particularly prevalent in industry. These processes produce MHP and MSP, which are intermediate products in the production of battery-grade nickel sulfate. MHP contained a lower grade of Ni than MSP. Leaching limonite ore typically yields a solution containing Ni, Co, Fe, and Mn. To produce high-purity MHP, precipitation removes Fe before leaching [68]. Leaching, purifying, and crystallizing the resulting MHPs produces battery-grade nickel sulfate. The HPAL route for treating limonite ore, followed by impurity removal using limestone to produce MHP intermediate, which is then re-leached with sulfuric acid and purified through solvent extraction, is a common process in Australia and Brazil. However, due to differences in ore grade and mineralogy, this process in Australia results in emissions of 27.54 kg CO₂-eq, with 37.9% of the emissions attributed to the electricity mix. In contrast, the same process in Brazil generates 12.67 kg CO₂-eq [45].

Thermal decomposition and reduction of limonite ore enhance the hydrometallurgical reduction process [69]. The Caron process, which uses a combination of pyrometallurgy and hydrometallurgy, processes limonite ores. During this process, the ore underwent reductive roasting followed by ammonia leaching [70]. The use of natural gas in this process can result in a high GWP of 13.8 kg CO₂-eq [45]. A new process developed by Oliveira et al. integrated pyrometallurgy and biohydrometallurgy to extract 92% nickel and 35% cobalt using only 35 g of sulfuric acid per kg of ore, a significant reduction compared to the HPAL method, which requires nearly 400 g of sulfuric acid per kg of ore. The process uses the bacterium *Acidithiobacillus ferrooxidans* for bioleaching at 32°C. This method increased the Eh value, which triggered the leaching of Ni- and Co-containing metallic phases. In contrast, a high pH value led to the precipitation of ferric iron as jarosite, which helped reduce acid consumption [69].

Advancements in bioleaching technologies can make hydrometallurgical processes greener by lowering sulfuric acid consumption, which has a significant environmental impact. Researchers have extensively studied bioleaching for limonitic laterite ore. Certain acidophilic, chemolithoautotrophic bacteria reduce ferric iron while metabolically oxidizing elemental sulfur to sulfuric acid under anaerobic and aerobic conditions. Bioleaching offers advantages such as low energy and acid consumption, making it a more sustainable extraction method [71].

Pyrometallurgical methods, such as smelting or sintering, can also be used to treat limonite ores. However, sintering in blast furnaces requires high fuel consumption and low productivity. The blast furnace smelting process is energy-intensive and requires a large amount of coke. A more environmentally friendly alternative, selective reductive smelting, such as the rotary kiln electric furnace (RK-EF) process, is widely used to treat Ni ore. However, owing to the high iron-to-nickel ratio of limonite ore, RK-EF produces low-grade ferronickel [72]. To produce battery-grade Ni, Ni pig iron, or Ni must be further converted into Ni matte. Hence, owing to the increased processing and high energy consumption of pyrometallurgy, hydrometallurgy, or a combination of pyro- and hydrometallurgy methods are used.

3.3.2.2.2. Saprolite ore

Saprolite ores have higher magnesium-silicon oxide and Ni contents than limonite ores. However, the high Mg content creates challenges for hydrometallurgical treatment, leading to a preference for pyrometallurgical techniques for treating saprolite ores. Several pyrometallurgical technologies can be employed for this purpose, such as rotary kiln-electric furnace (RK-EF), the Vaniukov process, direct reduction, submerged electric arc furnace, rotary hearth furnace, and smelting in a blast furnace. RK-EF-based treatment processes can have a high climate change impact, with emissions reaching approximately 86.4 kg CO₂ eq, especially in Indonesia [45]. Smelting saprolite ore produces ferronickel, which is further processed into nickel matte and treated to produce battery-grade nickel sulfate.

The new bloomery process melts ore, flux additives such as limestone, and reducers (brown coal, anthracite, or coal) in a rotary tube kiln at 1300–1400 °C to form granules. The granules were then cooled and separated by screening and magnetic separation [73]. Although this process produces granulated metallic Ni, further processing is required to obtain battery-grade nickel sulfate. Additionally, using coal for energy in this process produces high CO₂ emissions, reducing its environmental sustainability.

The high MgO/SiO₂ ratio in the saprolite ore leads to the formation of iron-nickel-magnesium silicates, which are difficult to reduce and result in low nickel recovery [74]. Reductive roasting of saprolite ore also faces a few challenges, such as (1) the liberation of Ni and Fe from their host minerals, (2) the sufficient reduction of Ni and Fe oxides, and (3) the growth of ferronickel particles. Therefore, additives and high-temperature reduction processes are required to overcome these limitations. For example, Wang et al. added waste red mud containing CaO and Na₂O to saprolite ore and subjected it to reductive roasting at 1200–1300°C for 60 min, followed by magnetic separation. This approach resulted in a nickel recovery of 95.25% owing to the reaction of CaO and Na₂O with SiO₂, leading to the formation of augite [75]. Other additives, such as sulfur, have also been investigated to improve reductive roasting [76]. Sari Y et al. investigated a more sustainable approach for the reductive roasting of saprolite ore, utilizing palm kernel shell charcoal (PSC) as a biomass reductant due to its high specific surface area of 217.2 m²g⁻¹. However, the nickel recovery rate from this method was relatively low at 27.4%. The addition of suitable

additives could potentially improve nickel recovery [77]. Use of biomass makes the process eco-friendly; however, a low recovery rate is the bottleneck for this process, which requires further research.

Environmental concerns have prompted the exploration of hydrometallurgical treatments of saprolite ores. In one study, column leaching of low-grade saprolitic ore achieved 72.5% Ni and 47.4% Co recovery using a solution containing $147 \text{ gL}^{-1} \text{ H}_2\text{SO}_4$ and $20 \text{ gL}^{-1} \text{ Na}_2\text{SO}_3$ [78]. Zhao Y investigated a microwave-assisted hydrothermal leaching process. In this method, saprolite ore is first roasted at 300°C , followed by leaching in a microwave-heated autoclave. Adding NaOH to the solution precipitated $\text{Fe}(\text{OH})_3$, removing Fe, and formed a mixed precipitate of $\text{Ni}(\text{OH})_2$ and $\text{Co}(\text{OH})_2$. This process achieved recoveries of 77.29% for Ni and 65.87% for Co [79]. Although these processes emit low GHGs and are considered environmentally friendly, their low recovery rates produce more waste. Increasing recovery from these processes is essential to consider them eco-friendly truly. Further purification of these mixed hydroxides will produce battery-grade nickel. Li G et al. developed a novel zero-waste approach using a combined hydrometallurgical process, removing 92% of iron as iron phosphate while recovering 98% of nickel and 100% of cobalt. The addition of phosphoric acid and sodium hydroxide played a crucial role in minimizing iron losses during the process [80]. Employment of such techniques that reduce waste is another way of developing sustainable solutions.

3.4. Environmental impact

The introduction of EVs aims to address global warming, making it crucial to evaluate the environmental impact of EVs throughout their lifecycle. Unlike vehicles with internal combustion engines, EVs produce minimal GHG emissions during the use phase because there is no fuel combustion. However, battery production contributes significantly to GHG emissions, highlighting the importance of assessing the environmental impacts at each stage of battery production. Currently, LCA is widely used in both academia and industry to evaluate the environmental impacts of processes, methods, and products. Positive electrode production has been identified as a major contributor to GHG emissions, with the mining and extraction of key minerals, particularly nickel and alumina, accounting for a significant portion. Emissions associated with these processes range from 38 to 62 kg $\text{CO}_2\text{-eq}$, depending on the geographic location. This variation primarily attributes to the regional differences in energy sources used for heat and electricity, which play a crucial role in determining the overall environmental impact. Battery chemistry significantly influences the overall GHG emissions associated with LIB production. In nickel-based chemistries, for example, cathode manufacturing contributes approximately 60% of the total emissions, with CAM synthesis alone accounting for nearly 44% of this impact [81]. Similarly, a comparative study revealed that raw material processing, including the production of battery-grade salts, constitutes around 40% of the GHG emissions in lithium iron phosphate (LFP) batteries (42 kg $\text{CO}_2\text{-eq/kWh}$), and as much as 55% in NMC 811 cells (52 kg $\text{CO}_2\text{-eq/kWh}$) [82]. These findings underscore the urgent need to improve the environmental performance of raw material production processes, particularly the conversion of minerals into battery-grade materials.

Table 1 presents an analysis of the mining of minerals, such as Li and Ni. Environmental parameters vary significantly across countries because of differences in mineral extraction technologies, electricity sources, and final products. For lithium, extraction from brine generally results in lower GHG emissions compared to extraction from spodumene ore. However, this

method is associated with significantly higher water consumption. Furthermore, the environmental impact varies significantly depending on the final lithium product, either LiOH or Li₂CO₃.

Table 1: LCA of mineral extraction of ores

<i>Mineral</i>	<i>Ores</i>	<i>Processing</i>	<i>Functional unit</i>	<i>kg CO₂ eq</i>	<i>Energy consumption (MJ)</i>	<i>Water consumption (m³)</i>	<i>Location</i>	<i>Ref</i>
<i>Lithium</i>	Spodumene	Roasting-leaching	1 tonne LiOH	15700	187200	3.4	Australia	[83]
	Spodumene	Roasting-leaching with Na ₂ CO ₃	1 tonne Li ₂ CO ₃	20400	218000	77	Australia	
	Brine	-	1 tonne LiOH	6900-7300	76600-82900	31-50	Salar de Atacama	
	Brine	-	1 tonne Li ₂ CO ₃	2700-3100	30000-36000	15.5-32.8	Salar de Atacama	
	Brine	Conventional method	1 tonne LCE	6170	-	70.3	Salar de Atacama	[27]
	Brine	Conventional method	1 tonne LCE	9300	-	62.1	Salar de Cauchari	
<i>Nickel</i>	Spodumene	-	1 tonne LCE	29200		99.4	Australia and China	
	Ni-Co-Cu	-	1 kg NiSO ₄	3.92		0.045	Canada and Norway	[84]
	Ni-Co-Cu	-	1 kg nickel	11.19		0.053		[85]
	Laterite and Sulphide	Pyrometallurgy and Hydrometallurgy	1 kg nickel	7.64	147		Global except China	[86]
	Low-grade limonitic laterites	HPAL	1 kg of nickel in NiSO ₄ ·6H ₂ O	36.8	385		Indonesia and China	[38]

3.5. Concluding remarks and perspectives

This chapter provides a concise yet comprehensive overview of two critical minerals, lithium and nickel, that are indispensable for powering LIBs. The geographically concentrated nature of their deposits, such as the Lithium Triangle (comprising Bolivia, Chile, and Argentina) for lithium and countries like Indonesia and Australia for nickel, poses significant supply chain risks. These geographic imbalances contribute to market volatility and underscore the urgent need to develop localized, geopolitically stable supply chains for battery-grade salts, ensuring uninterrupted material availability.

Spodumene ores and continental brines are the primary sources for lithium extraction. Each route has distinct advantages and limitations. Spodumene offers higher lithium concentrations but requires energy-intensive processing and results in elevated GHG emissions. In contrast, brine-based extraction is less carbon-intensive but suffers from high water consumption. However, recent advancements in DLE technologies have significantly improved the environmental sustainability of brine processing. Owing to these benefits, an increasing number of companies are adopting DLE methods for commercial use.

Nickel sulfide and laterite ores differ significantly in their mineralogy and, consequently, require different processing methods. Although industries historically preferred sulfide ores for their ease of processing and valuable by-products, the depletion of high-grade reserves has made extraction more challenging and energy-intensive. Traditional smelting methods generate substantial SO₂ emissions and can result in significant material losses. As a result, alternative approaches such as roasting-leaching have gained more prominence. Hydrometallurgical or combined pyro- and hydrometallurgical techniques are used to process laterite ores. Among emerging technologies, bioleaching has garnered interest due to its environmental benefits. However, its commercial application remains limited due to low extraction efficiency. Enhancing the performance and scalability of bioleaching could make it a viable, eco-friendly solution for nickel recovery.

Since mineral extraction stages contribute the most to the total GHG emissions in the lithium-ion battery value chain, it is imperative to focus on developing more environmentally responsible extraction methods. Innovations, such as direct ore-to-battery-grade salt conversion processes, offer promising opportunities to reduce environmental impact while enhancing economic feasibility. Future work should focus on developing a zero-waste process, potentially by utilizing by-products from other processes, thereby promoting a circular economy. In parallel, the formulation and enforcement of robust policies promoting sustainable extraction practices are essential to minimize ecological degradation and ensure a stable, long-term supply of critical materials.

References

1. M. Reich and A. C. Simon, "Critical Minerals," *Annu Rev Earth Planet Sci*, vol. 53, no. 1, pp. 141–168, May 2025, doi: 10.1146/ANNUREV-EARTH-040523-023316/CITE/REFWORKS.
2. "Critical Minerals Data Explorer – Data Tools - IEA." Accessed: Jul. 02, 2025. [Online]. Available: <https://www.iea.org/data-and-statistics/data-tools/critical-minerals-data-explorer>
3. "Sustainable Critical Minerals Alliance – Policies - IEA." Accessed: Jul. 02, 2025. [Online]. Available: <https://www.iea.org/policies/17635-sustainable-critical-minerals-alliance?s=1>
4. "Mineral commodity summaries 2024," 2024. doi: 10.3133/mcs2024.
5. R. J. Bowell, L. Lagos, C. R. de los Hoyos, and J. Declercq, "Classification and Characteristics of Natural Lithium Resources," *Elements*, vol. 16, no. 4, pp. 259–264, Aug. 2020, doi: 10.2138/GSELEMENTS.16.4.259.
6. S. E. Kesler, P. W. Gruber, P. A. Medina, G. A. Keoleian, M. P. Everson, and T. J. Wallington, "Global lithium resources: Relative importance of pegmatite, brine and other deposits," *Ore Geol Rev*, vol. 48, pp. 55–69, Oct. 2012, doi: 10.1016/J.OREGEOREV.2012.05.006.
7. "Essential Insights: Lithium Costs & Margins | S&P Global Market Intelligence." Accessed: Sep. 08, 2024. [Online]. Available: <https://pages.marketintelligence.spglobal.com/Lithium-brine-vs-hard-rock-demo-confirmation-MJ-ad.html>
8. F. Aguirre B, "The lithium triangle – the importance of Bolivia," *Journal of Energy & Natural Resources Law*, vol. 40, no. 2, pp. 183–202, Apr. 2022, doi: 10.1080/02646811.2021.1930708.
9. M. S. A. Chaudary, "Lithium dreams, local struggles: Navigating the geopolitics and socio-ecological costs of a low-carbon future," *Energy Res Soc Sci*, vol. 121, p. 103952, Mar. 2025, doi: 10.1016/J.ERSS.2025.103952.
10. J. S. Gutierrez, J. N. Moore, J. P. Donnelly, C. Dorador, J. G. Navedo, and N. R. Senner, "Climate change and lithium mining influence flamingo abundance in the Lithium Triangle," *Proceedings of the Royal Society B*, vol. 289, no. 1970, 2022, doi: 10.1098/RSPB.2021.2388.
11. "Nickel in batteries." Accessed: Sep. 08, 2024. [Online]. Available: <https://nickelinstitute.org/en/nickel-applications/nickel-in-batteries/>
12. G. M. Mudd, "Global trends and environmental issues in nickel mining: Sulfides versus laterites," *Ore Geol Rev*, vol. 38, no. 1–2, pp. 9–26, Oct. 2010, doi: 10.1016/J.OREGEOREV.2010.05.003.
13. T. Schmidt, M. Buchert, and L. Schebek, "Investigation of the primary production routes of nickel and cobalt products used for Li-ion batteries," *Resour Conserv Recycl*, vol. 112, pp. 107–122, Sep. 2016, doi: 10.1016/J.RESCONREC.2016.04.017.
14. "Nickel industry - Part 1 - Processing nickel laterites and sulfides." Accessed: Sep. 08, 2024. [Online]. Available: <https://nickelinstitute.org/en/blog/2024/may/nickel-industry-part-1-processing-nickel-laterites-and-sulfides/>
15. "Nickel recycling." Accessed: Sep. 12, 2024. [Online]. Available: <https://nickelinstitute.org/en/policy/nickel-life-cycle-management/nickel-recycling/>
16. "Struggling nickel miners press for alternative pricing mechanisms | Benchmark Source." Accessed: Sep. 12, 2024. [Online]. Available:

- <https://source.benchmarkminerals.com/article/struggling-nickel-miners-press-for-alternative-pricing-mechanisms>
17. D. I. Groves, D. Müller, M. Santosh, and C. X. Yang, “The heterogeneous distribution of critical metal mineral resources: An impending geopolitical issue,” *Geosystems and Geoenvironment*, vol. 4, no. 1, p. 100288, Feb. 2025, doi: 10.1016/J.GEOGEO.2024.100288.
 18. X. Q. Wang, T. Wu, H. Zhong, and C. W. Su, “Bubble behaviors in nickel price: What roles do geopolitical risk and speculation play?,” *Resources Policy*, vol. 83, p. 103707, Jun. 2023, doi: 10.1016/J.RESOURPOL.2023.103707.
 19. “Nickel industry margins surged in 2021 amid stronger nickel prices | S&P Global Market Intelligence.” Accessed: Sep. 12, 2024. [Online]. Available: <https://www.spglobal.com/marketintelligence/en/news-insights/research/nickel-industry-margins-surged-in-2021-amid-stronger-nickel-prices>
 20. H. Liu and G. Azimi, “Production of Battery Grade Lithium Hydroxide Monohydrate Using Barium Hydroxide Causticizing Agent,” *Resour Conserv Recycl*, vol. 179, p. 106115, Apr. 2022, doi: 10.1016/J.RESCONREC.2021.106115.
 21. S. Ahmed, A. K. Madikere Raghunatha Reddy, and K. Zaghib, “Transformations of Critical Lithium Ores to Battery-Grade Materials: From Mine to Precursors,” *Batteries 2024*, Vol. 10, Page 379, vol. 10, no. 11, p. 379, Oct. 2024, doi: 10.3390/BATTERIES10110379.
 22. J. Rioyo, S. Tuset, and R. Grau, “Lithium Extraction from Spodumene by the Traditional Sulfuric Acid Process: A Review,” *Mineral Processing and Extractive Metallurgy Review*, vol. 43, no. 1, pp. 97–106, Jan. 2022, doi: 10.1080/08827508.2020.1798234.
 23. A. Mas-Fons, R. Horta Arduin, P. Loubet, T. Pereira, A. M. Parvez, and G. Sonnemann, “Carbon and water footprint of battery-grade lithium from brine and spodumene: A simulation-based LCA,” *J Clean Prod*, vol. 452, p. 142108, May 2024, doi: 10.1016/J.JCLEPRO.2024.142108.
 24. G. D. Rosales, M. D. C. Ruiz, and M. H. Rodriguez, “Novel process for the extraction of lithium from β -spodumene by leaching with HF,” *Hydrometallurgy*, vol. 147–148, pp. 1–6, Aug. 2014, doi: 10.1016/J.HYDROMET.2014.04.009.
 25. S. Wang et al., “Direct Lithium Extraction from α -Spodumene through Solid-State Reactions for Sustainable Li_2CO_3 Production,” *Inorg Chem*, vol. 63, no. 29, pp. 13576–13584, Jul. 2024, doi: 10.1021/ACS.INORGCHEM.4C01722/ASSET/IMAGES/LARGE/IC4C01722_0006.JPG.
 26. H. Zhang et al., “Direct extraction of lithium from ores by electrochemical leaching,” *Nature Communications* 2024 15:1, vol. 15, no. 1, pp. 1–11, Jun. 2024, doi: 10.1038/s41467-024-48867-0.
 27. S. Mousavinezhad, S. Nili, A. Fahimi, and E. Vahidi, “Environmental impact assessment of direct lithium extraction from brine resources: Global warming potential, land use, water consumption, and charting sustainable scenarios,” *Resour Conserv Recycl*, vol. 205, p. 107583, Jun. 2024, doi: 10.1016/J.RESCONREC.2024.107583.
 28. J. Farahbakhsh et al., “Direct lithium extraction: A new paradigm for lithium production and resource utilization,” *Desalination*, vol. 575, p. 117249, Apr. 2024, doi: 10.1016/J.DESAL.2023.117249.
 29. “What Is Direct Lithium Extraction? - Lithium Harvest.” Accessed: Jul. 05, 2025. [Online]. Available: <https://lithiumharvest.com/knowledge/lithium-extraction/what-is-direct-lithium-extraction/>

30. Y. Boroumand and A. Razmjou, "Adsorption-type aluminium-based direct lithium extraction: The effect of heat, salinity and lithium content," *Desalination*, vol. 577, p. 117406, May 2024, doi: 10.1016/J.DESAL.2024.117406.
31. F. Arroyo, J. Morillo, J. Usero, D. Rosado, and H. El Bakouri, "Lithium recovery from desalination brines using specific ion-exchange resins," *Desalination*, vol. 468, p. 114073, Oct. 2019, doi: 10.1016/J.DESAL.2019.114073.
32. S. Yang, Y. Wang, H. Pan, P. He, and H. Zhou, "Lithium extraction from low-quality brines," *Nature*, vol. 636, no. 8042, pp. 309–321, Dec. 2024, doi: 10.1038/S41586-024-08117-1;SUBJMETA=4062,4072,4077,638,639,675;KWRD=DEVICES+FOR+ENERGY+HARVESTING,ENERGY.
33. Z. H. Foo and J. H. Lienhard, "Emerging membrane technologies for sustainable lithium extraction from brines and leachates: Innovations, challenges, and industrial scalability," *Desalination*, vol. 598, p. 118411, Apr. 2025, doi: 10.1016/J.DESAL.2024.118411.
34. H. Li et al., "Nanofiltration membrane with crown ether as exclusive Li⁺ transport channels achieving efficient extraction of lithium from salt lake brine," *Chemical Engineering Journal*, vol. 438, p. 135658, Jun. 2022, doi: 10.1016/J.CEJ.2022.135658.
35. J. Park et al., "Electrochemical Direct Lithium Extraction: A Review of Electrodialysis and Capacitive Deionization Technologies," *Resources* 2025, Vol. 14, Page 27, vol. 14, no. 2, p. 27, Feb. 2025, doi: 10.3390/RESOURCES14020027.
36. W. R. Torres, C. H. Díaz Nieto, A. PrévotEAU, K. Rabaey, and V. Flexer, "Lithium carbonate recovery from brines using membrane electrolysis," *J Memb Sci*, vol. 615, p. 118416, Dec. 2020, doi: 10.1016/J.MEMSCI.2020.118416.
37. S. Nikfar, A. Fahimi, and E. Vahidi, "Unlocking sustainable lithium: A comparative life cycle assessment of innovative extraction methods from brine," *Resour Conserv Recycl*, vol. 212, p. 107977, Jan. 2025, doi: 10.1016/J.RESCONREC.2024.107977.
38. G. Bartzas and K. Komnitsas, "Cradle to gate life-cycle assessment of battery grade nickel sulphate production through high-pressure acid leaching," *Science of The Total Environment*, vol. 952, p. 175902, Nov. 2024, doi: 10.1016/J.SCITOTENV.2024.175902.
39. J. Lu, S. Liu, J. Shangguan, W. Du, F. Pan, and S. Yang, "The effect of sodium sulphate on the hydrogen reduction process of nickel laterite ore," *Miner Eng*, vol. 49, pp. 154–164, Aug. 2013, doi: 10.1016/J.MINENG.2013.05.023.
40. V. Kirjavainen and K. Heiskanen, "Some factors that affect beneficiation of sulphide nickel–copper ores," *Miner Eng*, vol. 20, no. 7, pp. 629–633, Jun. 2007, doi: 10.1016/J.MINENG.2007.01.001.
41. K. Zhao et al., "Effect of a novel phosphate on the flotation of serpentine-containing copper-nickel sulfide ore," *Miner Eng*, vol. 150, p. 106276, May 2020, doi: 10.1016/J.MINENG.2020.106276.
42. F. Wang, F. Liu, R. Elliott, S. Rezaei, L. T. Khajavi, and M. Barati, "Solid State Extraction of Nickel from Nickel Sulfide Concentrates," *J Alloys Compd*, vol. 822, p. 153582, May 2020, doi: 10.1016/J.JALLCOM.2019.153582.
43. "Nickel Smelting and Refining", Accessed: Dec. 20, 2024. [Online]. Available: <http://www.ifc.org/ifcext/enviro.nsf/Content/EnvironmentalGuidelines>
44. W. Sun, X. Li, R. Liu, Q. Zhai, and J. Li, "Recovery of Valuable Metals from Nickel Smelting Slag Based on Reduction and Sulfurization Modification," *Minerals* 2021, Vol. 11, Page 1022, vol. 11, no. 9, p. 1022, Sep. 2021, doi: 10.3390/MIN11091022.

45. F. Valencia, M. Rabbani, A. Fahimi, and E. Vahidi, "Assessing the environmental burden of nickel sulfate for batteries: A life cycle perspective," *Resour Conserv Recycl*, vol. 215, p. 108130, Apr. 2025, doi: 10.1016/J.RESCONREC.2025.108130.
46. W. Xiao, X. Chen, X. Liu, Z. Zhao, and Y. Li, "A method for extracting valuable metals from low nickel matte by non-oxidative leaching with H₂SO₄," *Sep Purif Technol*, vol. 270, p. 118789, Sep. 2021, doi: 10.1016/J.SEPPUR.2021.118789.
47. M. H. Morcali, L. T. Khajavi, S. Aktas, and D. B. Dreisinger, "Oxidative dissolution of nickel matte in dilute sulfuric acid solutions," *Hydrometallurgy*, vol. 185, pp. 257–265, May 2019, doi: 10.1016/J.HYDROMET.2019.03.010.
48. J. Chang et al., "Efficient reductive leaching of valuable elements from oxygen pressure leaching residue of high-grade nickel matte within sulfur dioxide," *Chemical Engineering Journal*, vol. 483, p. 149295, Mar. 2024, doi: 10.1016/J.CEJ.2024.149295.
49. M. de Fátima da Silva, M. R. de Sousa Oliveira, I. D. dos Santos, P. Radino-Rouse, and M. B. Mansur, "Iron Precipitation Strategies from Nickel Laterite Ore Sulfuric Acid Leach Liquor," *Mineral Processing and Extractive Metallurgy Review*, vol. 43, no. 1, pp. 28–39, Jan. 2022, doi: 10.1080/08827508.2020.1809392.
50. K. C. Sole, "The Evolution of Cobalt–Nickel Separation and Purification Technologies: Fifty Years of Solvent Extraction and Ion Exchange," pp. 1167–1191, 2018, doi: 10.1007/978-3-319-95022-8_95.
51. F. Cui et al., "Synchronous extractions of nickel, copper, and cobalt by selective chlorinating roasting and water leaching to low-grade nickel-copper matte," *Sep Purif Technol*, vol. 195, pp. 149–162, Apr. 2018, doi: 10.1016/J.SEPPUR.2017.11.071.
52. F. Cui, W. Mu, S. Wang, Q. Xu, Y. Zhai, and S. Luo, "Controllable Phase Transformation in Extracting Valuable Metals from Chinese Low-Grade Nickel Sulphide Ore," *JOM*, vol. 69, no. 10, pp. 1925–1931, Oct. 2017, doi: 10.1007/S11837-017-2405-8/FIGURES/4.
53. W. Mu, R. Yang, J. Meng, M. Li, X. Lei, and S. Luo, "Eco-friendly and selective separation of nickel and copper from low-grade nickel sulfide ore via sulfur fixation roasting with MgCO₃ followed by sulfuric acid leaching," *Sep Purif Technol*, vol. 343, p. 127094, Sep. 2024, doi: 10.1016/J.SEPPUR.2024.127094.
54. X. Xu et al., "Direct Extraction of Nickel and Copper from Low-Grade Nickel Sulfide Ore by Chlorination Roasting with Mixed MgCl₂·6H₂O and NaCl," *JOM*, vol. 74, no. 5, pp. 1989–1999, May 2022, doi: 10.1007/S11837-021-05122-X/TABLES/2.
55. K. Meng, Z. Wang, H. Guo, X. Li, and J. Wang, "A compact process to prepare LiNi_{0.8}Co_{0.1}Mn_{0.1}O₂ cathode material from nickel-copper sulfide ore," *Hydrometallurgy*, vol. 174, pp. 1–9, Dec. 2017, doi: 10.1016/J.HYDROMET.2017.09.010.
56. P. Kinnunen, T. Riihimäki, K. Kinnunen, M. Salo, T. Heikola, and J. Mäkinen, "Process Design for Direct Production of Battery Grade Nickel Sulfate," *Journal of Sustainable Metallurgy*, pp. 1–11, Jun. 2024, doi: 10.1007/S40831-024-00864-9/TABLES/5.
57. S. Farrokhpay and L. Filippov, "Challenges in processing nickel laterite ores by flotation," *Int J Miner Process*, vol. 151, pp. 59–67, Jun. 2016, doi: 10.1016/J.MINPRO.2016.04.007.
58. C. Li, W. Liu, F. Jiao, T. Zhang, J. Han, and W. Qin, "Intensifying the selective sulfidation of nickel in saprolitic laterites by sodium salts," *Miner Eng*, vol. 218, p. 109022, Nov. 2024, doi: 10.1016/J.MINENG.2024.109022.
59. V. N. Khabashesku, F. Bella, P. Kalungi, Z. Yao, and H. Huang, "Aspects of Nickel, Cobalt and Lithium, the Three Key Elements for Li-Ion Batteries: An Overview on Resources, Demands, and Production," *Materials* 2024, Vol. 17, Page 4389, vol. 17, no. 17, p. 4389, Sep. 2024, doi: 10.3390/MA17174389.

60. U. König and M. Panalytical, “Nickel Laterites—Mineralogical Monitoring for Grade Definition and Process Optimization,” *Minerals* 2021, Vol. 11, Page 1178, vol. 11, no. 11, p. 1178, Oct. 2021, doi: 10.3390/MIN11111178.
61. N. Pandey, S. K. Tripathy, S. K. Patra, and G. Jha, “Recent Progress in Hydrometallurgical Processing of Nickel Lateritic Ore,” *Transactions of the Indian Institute of Metals*, vol. 76, no. 1, pp. 11–30, Jan. 2023, doi: 10.1007/S12666-022-02706-2/TABLES/12.
62. A. Oxley, M. E. Smith, and O. Caceres, “Why heap leach nickel laterites?,” *Miner Eng*, vol. 88, pp. 53–60, Mar. 2016, doi: 10.1016/J.MINENG.2015.09.018.
63. A. L. A. Santos, E. M. A. Becheleni, P. R. M. Viana, R. M. Papini, F. P. C. Silvas, and S. D. F. Rocha, “Kinetics of Atmospheric Leaching from a Brazilian Nickel Laterite Ore Allied to Redox Potential Control,” *Min Metall Explor*, vol. 38, no. 1, pp. 187–201, Feb. 2021, doi: 10.1007/S42461-020-00310-W/TABLES/5.
64. Q. hua Tian, B. Dong, X. yi Guo, Q. ao Wang, Z. peng Xu, and D. Li, “Valuable metals substance flow analysis in high-pressure acid leaching process of laterites,” *J Cent South Univ*, vol. 30, no. 6, pp. 1776–1786, Jun. 2023, doi: 10.1007/S11771-023-5356-Y/METRICS.
65. X. Xiong et al., “Hydrometallurgical process and recovery of valuable elements for limonitic laterite: A review,” *Chin J Chem Eng*, vol. 73, pp. 189–201, Sep. 2024, doi: 10.1016/J.CJCHE.2024.05.011.
66. H. L. Carpen and E. C. Giese, “Enhancement of nickel laterite ore bioleaching by *Burkholderia* sp. using a factorial design,” *Appl Water Sci*, vol. 12, no. 8, pp. 1–8, Aug. 2022, doi: 10.1007/S13201-022-01704-5/TABLES/5.
67. M. H. Morcali, L. T. Khajavi, and D. B. Dreisinger, “Extraction of nickel and cobalt from nickeliferous limonitic laterite ore using borax containing slags,” *Int J Miner Process*, vol. 167, pp. 27–34, Oct. 2017, doi: 10.1016/J.MINPRO.2017.07.012.
68. N. Safitri, M. Z. Mubarak, R. Winarko, and Z. Tanlega, “Recovery of nickel and cobalt as MHP from limonitic ore leaching solution: Kinetics analysis and precipitate characterization,” *AIP Conf Proc*, vol. 1964, no. 1, p. 20030, May 2018, doi: 10.1063/1.5038312/974763.
69. V. de A. Oliveira, M. L. M. Rodrigues, and V. A. Leão, “Reduction roasting and bioleaching of a limonite ore,” *Hydrometallurgy*, vol. 200, p. 105554, Mar. 2021, doi: 10.1016/J.HYDROMET.2021.105554.
70. V. de A. Oliveira, C. G. Dos Santos, and E. de A. Brocchi, “Assessing the Influence of NaCl on the Reduction of a Siliceous Laterite Nickel Ore Under Caron Process Conditions,” *Metallurgical and Materials Transactions B: Process Metallurgy and Materials Processing Science*, vol. 50, no. 3, pp. 1309–1321, Jun. 2019, doi: 10.1007/S11663-019-01552-W/FIGURES/15.
71. S. Stanković et al., “Effect of mineralogy on Co and Ni extraction from Brazilian limonitic laterites via bioleaching and chemical leaching,” *Miner Eng*, vol. 184, p. 107604, Jun. 2022, doi: 10.1016/J.MINENG.2022.107604.
72. X. Wang et al., “Efficient Utilization of Limonite Nickel Laterite to Prepare Ferronickel by the Selective Reduction Smelting Process,” *Sustainability* 2023, Vol. 15, Page 7147, vol. 15, no. 9, p. 7147, Apr. 2023, doi: 10.3390/SU15097147.
73. G. B. Sadykhov, K. G. Anisovyan, D. Y. Kop’ev, K. B. Goncharov, and T. V. Olyunina, “Development of a Low-Temperature Bloomery Process for Nickel and Cobalt Recovery from Saprolite Ores,” *Russian Metallurgy (Metally)*, vol. 2020, no. 3, pp. 165–179, Mar. 2020, doi: 10.1134/S003602952003009X/TABLES/4.

74. F. Nurjaman, W. Astuti, F. Bahfie, and B. Suharno, "Study of selective reduction in lateritic nickel ore: Saprolite versus limonite," *Mater Today Proc*, vol. 44, pp. 1488–1494, Jan. 2021, doi: 10.1016/J.MATPR.2020.11.687.
75. X. ping Wang, T. chang Sun, J. Kou, Z. chun Li, and Y. Tian, "Feasibility of co-reduction roasting of a saprolitic laterite ore and waste red mud," *International Journal of Minerals, Metallurgy and Materials*, vol. 25, no. 6, pp. 591–597, Jun. 2018, doi: 10.1007/S12613-018-1606-7/METRICS.
76. A. Bunjaku, M. Kekkonen, S. Pekkarinen, and P. Taskinen, "Effect of sulphur addition on roasting reduction of saprolitic nickel ores in CO/CO₂ gas mixture," *Transactions of the Institutions of Mining and Metallurgy, Section C: Mineral Processing and Extractive Metallurgy*, vol. 122, no. 1, pp. 15–24, Mar. 2013, doi: 10.1179/1743285512Y.0000000025/ASSET/IMAGES/10.1179_1743285512Y.0000000025-IMG10008.PNG.
77. Y. Sari et al., "Nickel recovery in ferronickel concentrate by green selective reduction of nickel laterite," *Engineering Science and Technology, an International Journal*, vol. 57, p. 101798, Sep. 2024, doi: 10.1016/J.JESTCH.2024.101798.
78. K. Komnitsas, E. Petrakis, G. Bartzas, and V. Karmali, "Column leaching of low-grade saprolitic laterites and valorization of leaching residues," *Science of The Total Environment*, vol. 665, pp. 347–357, May 2019, doi: 10.1016/J.SCITOTENV.2019.01.381.
79. Y. Zhao et al., "Extraction and separation of nickel and cobalt from saprolite laterite ore by microwave-assisted hydrothermal leaching and chemical deposition," *International Journal of Minerals, Metallurgy and Materials*, vol. 20, no. 7, pp. 612–619, Jul. 2013, doi: 10.1007/S12613-013-0774-8/METRICS.
80. G. Li, Y. Chen, C. Mang, X. Teng, J. Luo, and M. Rao, "Near-Zero-Waste Approach for the Treatment of Saprolitic Nickel Laterite via a Combined Hydrometallurgical Process," *ACS Sustain Chem Eng*, vol. 11, no. 18, pp. 6856–6865, May 2023, doi: 10.1021/ACSSUSCHEMENG.2C05403/ASSET/IMAGES/LARGE/SC2C05403_0014.JPG.
81. Jorge A Llamas-Orozco, Fanran Meng, Gavin S Walker, Amir F N Abdul-Manan, Heather L MacLean, I Daniel Posen, Jon McKechnie, Estimating the environmental impacts of global lithium-ion battery supply chain: A temporal, geographical, and technological perspective, *PNAS Nexus*, Volume 2, Issue 11, November 2023, pgad361, <https://doi.org/10.1093/pnasnexus/pgad361>
82. Manjong, N. B., Usai, L., Orangi, S., Perez Clos, D., & Strømman, A. H. (2024). Exploring raw material contributions to the greenhouse gas emissions of lithium-ion battery production. *Journal of Energy Storage*, 100 (Part A), 113566. <https://doi.org/10.1016/j.est.2024.113566>
83. J. C. Kelly, M. Wang, Q. Dai, and O. Winjobi, "Energy, greenhouse gas, and water life cycle analysis of lithium carbonate and lithium hydroxide monohydrate from brine and ore resources and their use in lithium ion battery cathodes and lithium ion batteries," *Resources, Conservation & Recycling*, 2021, Vol. 174, Page 105762, Nov. 2021, doi: 10.1016/j.resconrec.2021.105762.
84. M. Bollwein, "Comparative life cycle assessment of prospective battery-grade material production in Norway," *NTNU Open*, 2022, [Online] Available: NTNU Open, Accessed: Jan. 12, 2025.

85. S. H. Farjana, N. Huda, and M. A. P. Mahmud, "Life cycle assessment of cobalt extraction process," *Journal of Sustainable Mining* 2019, Vol. 18, no. 3, pp. 150–161, Aug. 2019, doi: 10.1016/j.jsm.2019.03.002
86. M. Mistry, J. Gediga, and S. Boonzaier, "Life cycle assessment of nickel products," *The International Journal of Life Cycle Assessment* 2016, Vol. 21, no. 11, pp. 1559–1572, Mar. 2016, doi: 10.1007/s11367-016-1085-x.

Chapter 4 – Life cycle assessment of Lithium hydroxide

4.1. Introduction to Life Cycle Assessment

LCA is a standardized methodology defined by ISO 14040 for evaluating the environmental impacts associated with a product or service throughout its entire life cycle, from the extraction of natural resources, through production and use, to end-of-life treatment. One of the primary advantages of LCA is its ability to avoid shifting environmental burdens between life cycle stages, enabling the identification of the system's absolute environmental footprint under study [1]. LCA plays a crucial role in pinpointing the processes and activities with the highest environmental burdens and in evaluating alternative strategies that could reduce these impacts. LCA, in turn, supports the development of robust circular economy frameworks [2]. Importantly, LCA captures both direct and indirect environmental impacts, thereby providing a comprehensive perspective [3].

The methodology has four key phases:

- **Goal and Scope Definition** - This stage outlines the study's objective, its intended application, the rationale for conducting it, and the target audience. It also defines critical methodological parameters, such as the functional unit, system boundaries, allocation procedures, selected impact categories, LCIA models, and data quality requirements.
- **Life Cycle Inventory (LCI)** - This phase involves quantitative data collection to characterize the inputs (e.g., raw materials, energy) and outputs (e.g., products, emissions, waste) associated with the defined system.
- **Life Cycle Impact Assessment (LCIA)** - In this stage, impact assessment models translate the inventory data into environmental indicators across categories such as GWP, acidification, and eutrophication.
- **Interpretation** - The final phase involves analyzing the results to conclude, conducting sensitivity and uncertainty analyses, and ensuring the robustness and credibility of the findings [4].

LCA provides a robust and standardized framework for assessing the environmental impact of materials and processes throughout their entire life cycle. With the growing global demand for LIBs, it has become increasingly important to evaluate their associated environmental impacts. In the following sections, we focus on the environmental footprint of lithium hydroxide, a critical battery-grade salt essential for the synthesis of positive electrode active materials in LIBs.

4.2. Background

As discussed in Chapter 3, spodumene ore and lithium-rich brines are the primary sources of lithium. Brine deposits are predominantly located in the Lithium Triangle, comprising Chile, Argentina, and Bolivia, while Australia and China have significant spodumene deposits. In response to growing concerns regarding energy security, supply chain vulnerability, and the environmental impact of LIB production, there is an increasing urgency to develop a resilient lithium-ion supply chain within North America. This supply chain encompasses not only EV manufacturing but also cell fabrication and the production of critical raw materials [5].

Due to its abundant critical mineral reserves, technological advancements in manufacturing, and strong ESG (environmental, social, and governance) performance, Canada was ranked first in Bloomberg's 2024 Global Lithium-Ion Battery Supply Chain Ranking [6]. Strategic frameworks, such as the Canadian Critical Minerals Strategy, further underscore Canada's commitment to

supply chain development. Quebec, in particular, plays a pivotal role in advanced mining projects and processing infrastructure for essential LIB materials, including lithium, nickel, cobalt, graphite, and rare earth elements [7]. The diversity and scale of ongoing projects highlight Quebec's central position in the emerging North American EV supply chain.

To ensure the sustainability and environmental integrity of this supply chain, it is imperative to conduct a comprehensive environmental assessment. Researchers have performed several LCA studies on lithium extraction from brine and spodumene sources across different geographies. For instance, Chordia et al. evaluated and compared the environmental impacts of LHM production with varying lithium grades. For brines, the study comparedecoinvent data (0.15% Li) with field data from Kelly et al. for Salar de Atacama (0.17% Li), Salar de Cauchari (0.05% Li), and Salar de Maricunga (0.09% Li), reporting GWP of 5.5, 7.31, 15.6, and 17.3 t CO₂-eq, respectively, and water consumption values of 76.7, 80.2, 244, and 327 m³. For spodumene sources, they assessedecoinvent (1.86% Li) and Kelly et al. (0.80% Li) data from Australia, Canada (0.70% Li), and Finland (0.60% Li), reporting GWPs of 19.2, 18.5, 9.6, and 10.1 t CO₂-eq and water usage of 221, 120, 418, and 263 m³, respectively [8].

In another study, Mas-Fons et al. simulated lithium carbonate (Li₂CO₃) production from both brines (Salar de Atacama) and spodumene ore (mined in Australia and refined in China) across low-, medium-, and high-grade scenarios. The GWP ranged from 5.0 to 25.0 kg CO₂-eq per kg Li₂CO₃ for brines and from 17.1 to 22.3 kg CO₂-eq per kg Li₂CO₃ for spodumene. Water consumption for brine-based routes varied from 0.2 to 7.7 m³ per kg Li₂CO₃ (depending on accounting method), while spodumene-based production ranged from 0.2 to 0.5 m³ per kg Li₂CO₃. The study concluded that declining ore grades in brine operations cause a more significant increase in carbon footprint compared to spodumene [9]. Ibarra-Gutiérrez assessed Quebec's lithium production potential and estimated process-based emissions of 1,875 kg CO₂-eq per tonne of LHM from mining and electrochemical conversion. However, this study relied on emission factors rather than a complete LCA approach and therefore excluded indirect emissions and upstream impacts [10].

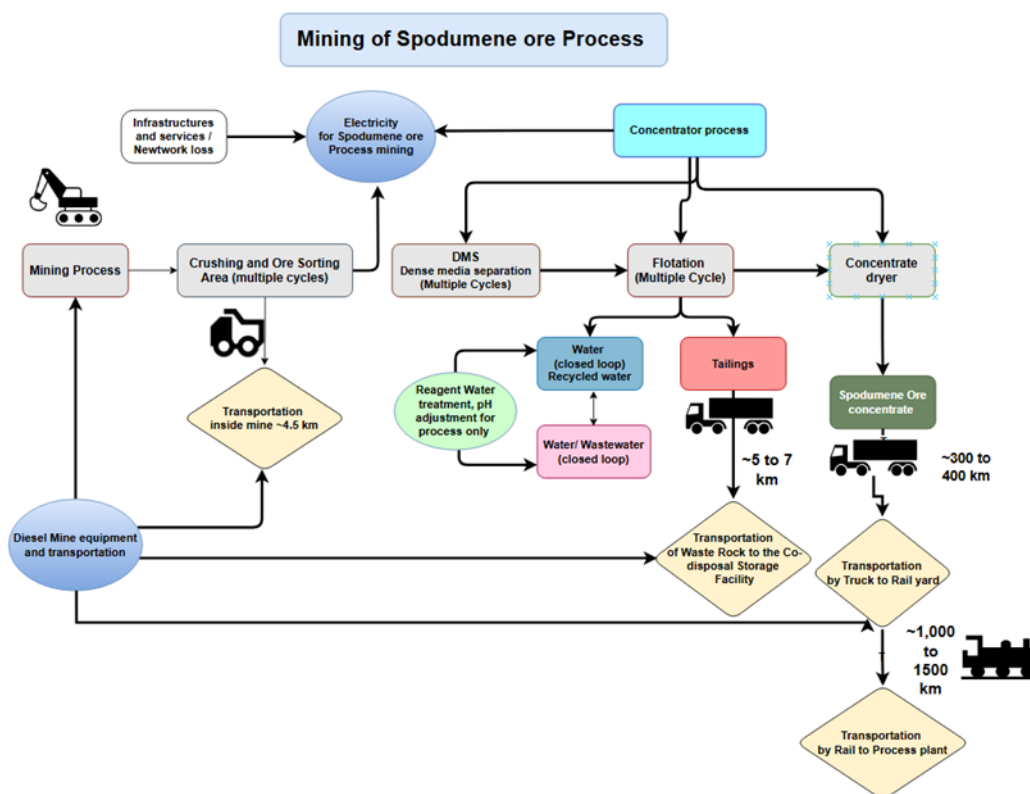
While extensive LCA studies exist for major producing regions, such as South America and Australia, a significant gap remains in the literature regarding Quebec's lithium production. This study aims to address that gap by conducting an LCA of lithium extraction and processing in Quebec. Our objective is not only to evaluate the environmental sustainability of Canada's lithium supply chain but also to highlight the emerging role of regions like Quebec, which, although not yet dominant producers, are crucial to the development of a North American lithium value chain.

4.3 Methods

4.3.1. Lithium processing in Quebec

Fig. 4 presents lithium ore processing (spodumene to concentrated lithium), and Fig. 5 shows the transformation from concentrated lithium to LHM. Québec spodumene contains approximately 1.2% lithium, and mining operations concentrate the ore to 5.5% at the site. Several beneficiation steps, including crushing, grinding, DMS, and flotation, to concentrate the lithium. In this LCA, the process assumes that concentrated spodumene is transported to another plant, where it converts into battery-grade LHM. The transformation process begins with the calcination of the ore, followed by roasting it with sulfuric acid and leaching. The leachate was further purified, neutralized, and crystallized to obtain battery-grade LHM. This process generates co-products,

Figure 4: Extraction of spodumene ore at mine and concentration of material in Québec



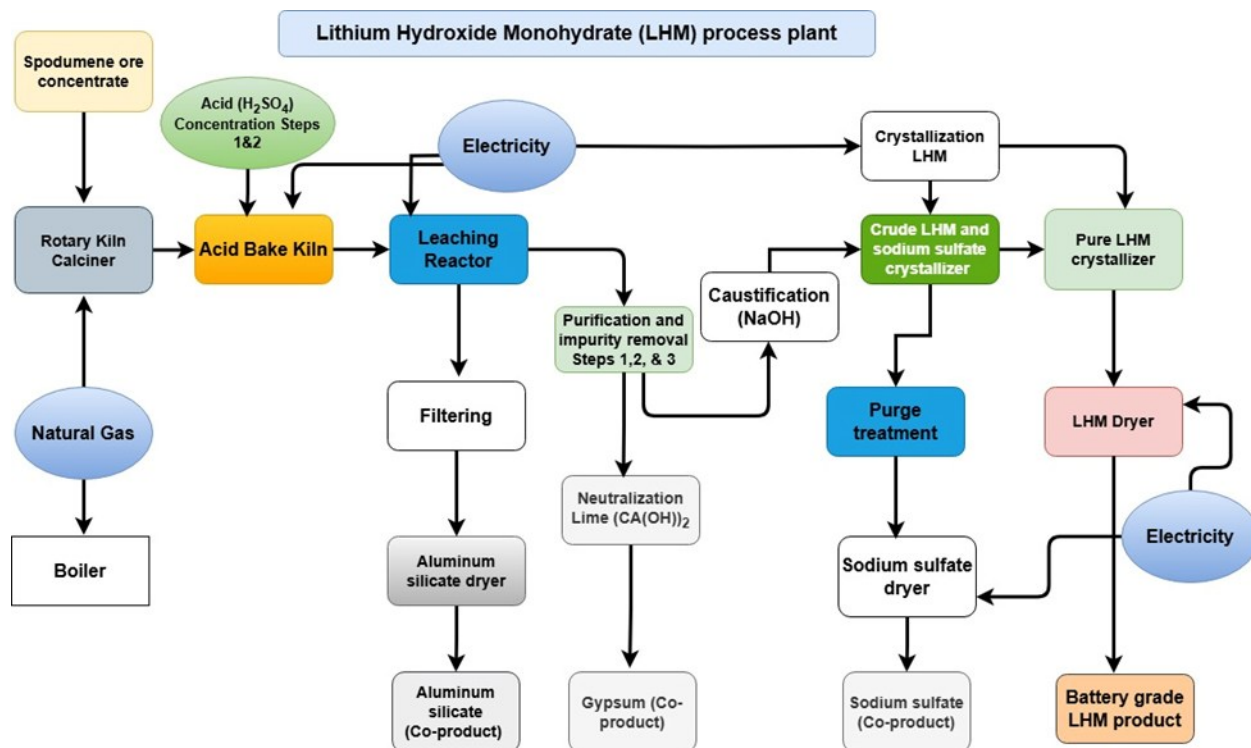


Figure 5: Concentrate to LHM Process Plant ($\text{LiOH} \cdot \text{H}_2\text{O}$)

4.3.2. Goal, Scope, and Inventory Analysis

This study conducts a cradle-to-gate LCA of battery-grade LHM produced from spodumene ore in Québec, Canada. The assessment follows the ISO 14040 standards for environmental management and LCA methodology. The primary objective is to quantify the environmental impacts associated with each stage of the extraction and processing chain, from raw material acquisition to the production of refined LHM suitable for LIB applications. The functional unit is defined as the production of 1 ton of battery-grade LHM, enabling comparability with prior studies and industrial benchmarks. Table 2 provides an overview of the goal, scope, and impact assessment of this research.

Table 2: Overview of the LCA parameters used in this study

Goal and Scope	Cradle-to-gate LCA of lithium extraction from spodumene
Functional Unit	1 ton of battery-grade LHM
Product Technology	Battery-grade LHM used in EV batteries
Foreground data	Spodumene mine and processing facility in Québec
Background database	Ecoinvent database (v3.8)

Cut-off criteria	No explicit cut-off criteria. All information on energy, materials, and emissions compiled from industry-specific technical documents
Impact Assessment Categories	Midpoint - Climate change and water scarcity

This study adopts a cradle-to-gate system boundary (as shown in Fig 6), encompassing the whole production chain from spodumene ore mining to the final crystallization and packaging of LHM. Specifically, the system includes

- Mining and beneficiation processes such as crushing, DMS, and flotation.
- Transport of spodumene concentrate to the process plant via truck and rail.
- LHM production processes include calcination, acid baking, leaching, purification, crystallization, drying, and packaging.
- All direct and indirect inputs, including electricity, diesel, gasoline, natural gas, reagents (e.g., H_2SO_4 , NaOH), and water usage.

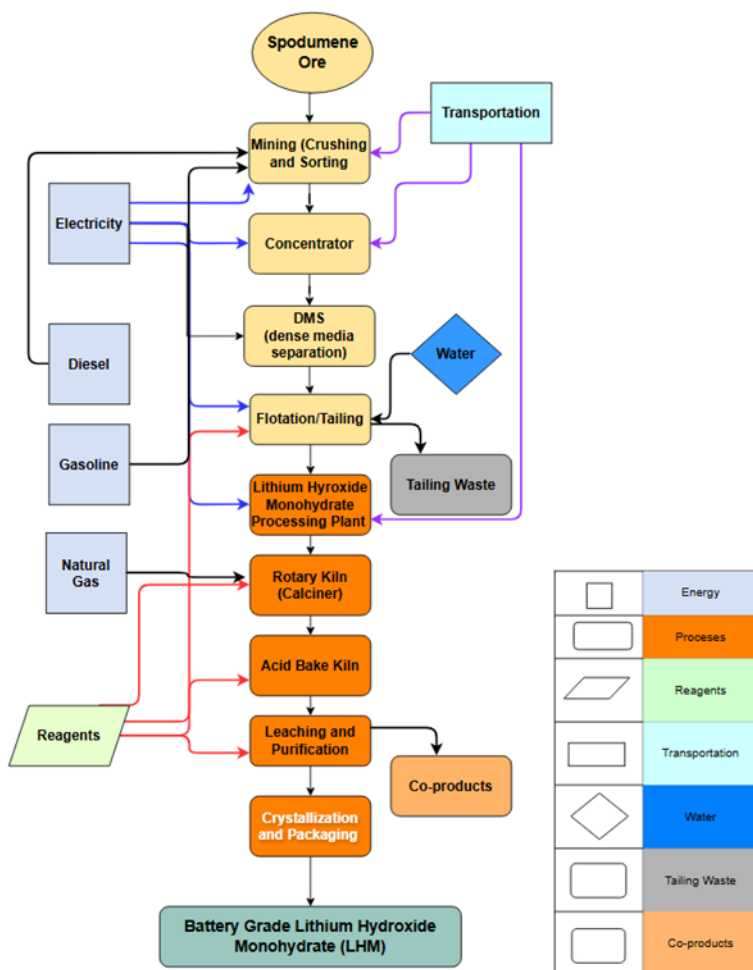


Figure 6: Process Tree (mine-to-material; LHM Québec Mine extraction/concentrator and process plant)

The study developed the LCI using both primary and secondary data. Foreground data were obtained directly from operational reports, feasibility studies, and environmental impact assessments (EIAs) for a spodumene mine and LHM processing facility in Québec [11-15]. These data included site-specific metrics for energy consumption, emissions, material flows, and waste generation. This study sourced background data from the Ecoinvent v3.8 database, ensuring alignment with global LCA datasets.

This study conducted inventory modeling using OpenLCA (version 2.3). It defined process flows for all relevant life cycle stages and validated the model prior to impact assessment. Table 3 lists the unit processes with their respective input and output flows as modeled in OpenLCA.

Table 3: List of all unit processes and their respective input and output flows, including raw materials, reagents, electricity, water, fuel, transportation, and waste streams as modelled in openLCA.

Unit process	Input flow	Output flow
LHM_Mining_Processes_at_Mine	Spodumene Diesel_fuel_drilling_blasting Electricity_mine_general_utilities Electricity_crushing_ore electricity_monitoring_station electricity_powerdistribution electricity_preconcentration Transport_in_mine Gasoline_light_duty_trucks	Mining_process non-sulfidic_tailings
LHM_Spodumene_ore_flotation	Mining_process tap water electricity_tailings_concentrator electricity_concentrator_reagents electricity_water_treatment neutralising_agent_NaOH	Spodumene_ore_flotation
LHM_Spodumene_ore_concentrator	Spodumene_ore_flotation electricity_dry_magnetic_separation electricity_wet_magnetic_separation electricity_concentrate_dryer	Spodumene_ore_concentrate
LHM_facility_wide_all_processes	Spodumene_ore_concentrate electricity_facility_wide natural_gas_kilns_boiler transport_between_mine_plant_rail transport_between_mine_plant_trucks	LHM_facility_wide_all_process

LHM_leaching	LHM_facility_wide_all_process sulfuric acid	LHM_leaching
LHM_purification	LHM_leaching	LHM_purification gypsum_co-product CO ₂ _emissions_from_Ca(O H) ₂ _production
LHM_crystallizer	LHM_purification neutralising_agent_NaOH	LHM_crystallizer Sodium_sulphate_co- product
Production_of_1ton_LHM	LHM_crystallizer	Production_of_1ton_LHM

The impact assessment was conducted using Impact World+ midpoint v2.0.1. Environmental indicators evaluated included midpoint categories: climate change or global warming potential (GWP, in kg CO₂-equivalent) and water scarcity (in m³ world-equivalent). This study employs an attributional LCA framework, which quantifies the direct environmental burdens associated with the defined production system, without accounting for market-mediated effects or consequential changes in system behaviour. This approach allows for a consistent, transparent evaluation of the environmental footprint of LHM production and enables identification of hotspots related to energy use, GHG emissions, and resource consumption.

4.4. Results and Discussion

4.4.1. Climate change

The impact assessment method, IMPACT World+ midpoint, provides two distinct evaluations for climate change: short-term and long-term. The short-term climate change indicator assesses near-term impacts (i.e., <100 years), making it relevant for evaluating progress toward short-term climate goals. In contrast, the long-term climate change indicator assesses the impacts of global temperature changes over a time horizon exceeding 100 years [16]. This distinction is crucial for greenhouse gases like methane, which have a short atmospheric lifetime and thus exhibit significantly different impacts depending on the assessment horizon. The GWP of methane over a 20-year time frame (GWP20) ranges from 84 to 87, whereas over a 100-year time frame (GWP100), it ranges from 28 to 36 [17].

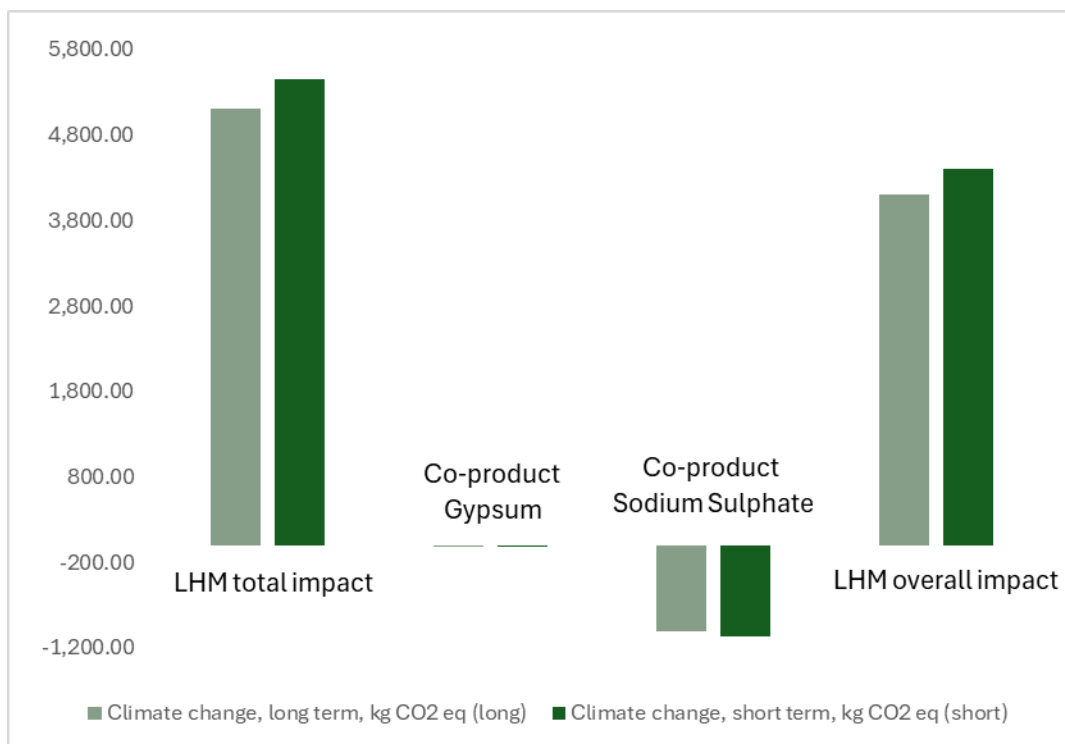


Figure 7: Climate change (short term and long term) impact of LHM production with co-product carbon offset credits

As shown in Fig. 7, the long-term climate change impact associated with LHM production is 5,110 kg CO₂-eq. However, this impact is reduced to 4,100.75 kg CO₂-eq when accounting for avoided burdens from co-products, primarily sodium sulfate (−1,007.95 kg CO₂-eq). Similarly, the short-term climate change impact begins at 5,459.10 kg CO₂-eq, which is reduced to 4,395.42 kg CO₂-eq after incorporating the climate benefits of co-products. The contribution from gypsum is nearly negligible, likely due to its low market value or limited demand. These results demonstrate that co-product recovery, particularly of high-impact offsets like sodium sulfate, can significantly reduce the overall climate change footprint of LHM production.

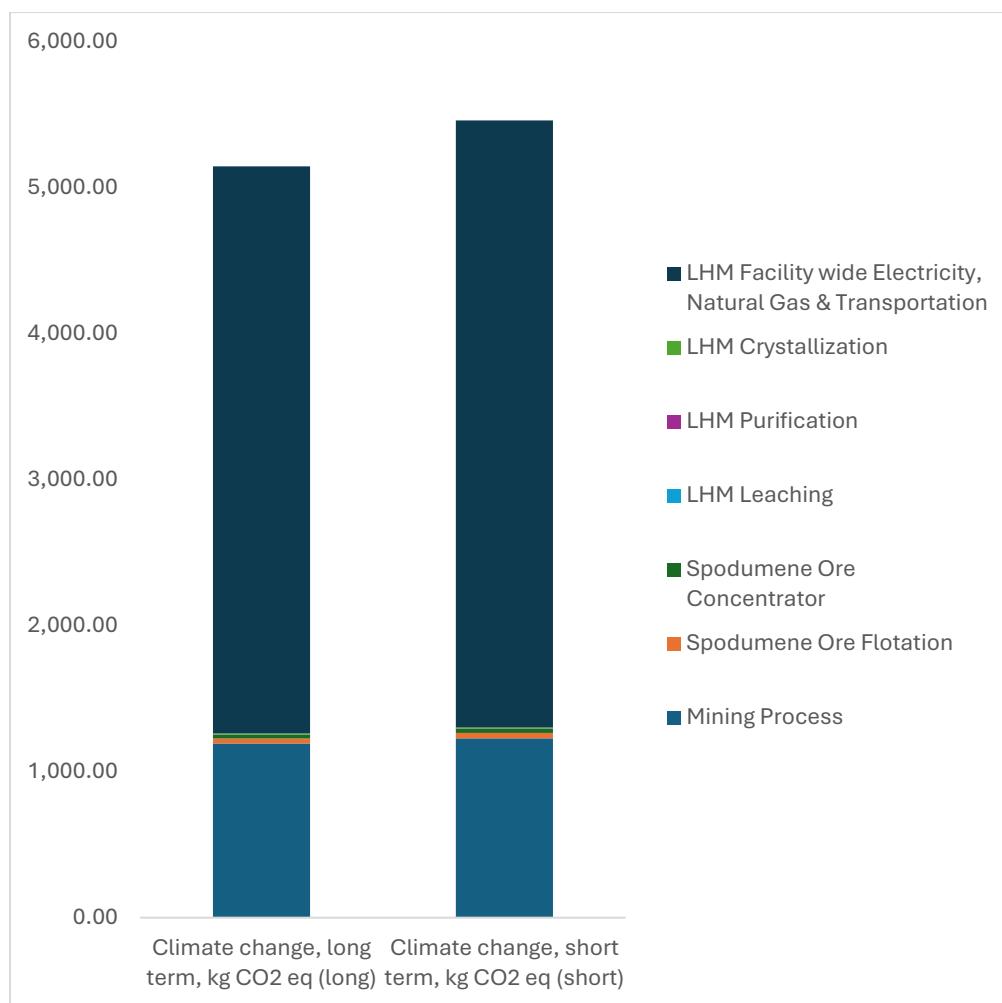


Figure 8: Breakdown of climate change impacts across individual unit processes in LHM production, identifying the primary contributors to overall CO₂-eq emissions

Fig. 8 highlights the climate change impact associated with various stages of LHM production in Québec without accounting for co-products. The most significant contributors to climate change impacts are facility-wide electricity consumption, natural gas use, and transportation, particularly the shipment of concentrate via truck and rail to the processing facility. While the GWP metric, whether short-term or long-term, affects absolute values, the relative contribution of each process stage remains the same.

The mining and ore processing stages collectively represent a notable share of the total emissions, with mining alone contributing 1,228.09 kg CO₂-eq (short-term). In contrast, the spodumene flotation and concentration stages contribute only 60.30 kg CO₂-eq (long-term) and 64.52 kg CO₂-eq (short-term), respectively. The reagents used in these processes have a negligible impact on the emissions. The LHM production facility-wide processes generate most emissions, accounting for 76% of the total impact. As detailed in Table 2, the unit process “LHM production facility-wide processes” encompasses key contributors such as electricity, natural gas, and transportation of the concentrate. The dominance of emissions from natural gas combustion, electricity use, and logistics underscores the importance of energy sourcing. Given that Québec’s

electricity mix is predominantly renewable, the associated emissions are significantly lower than in regions reliant on coal-based power.

4.4.2. Water scarcity

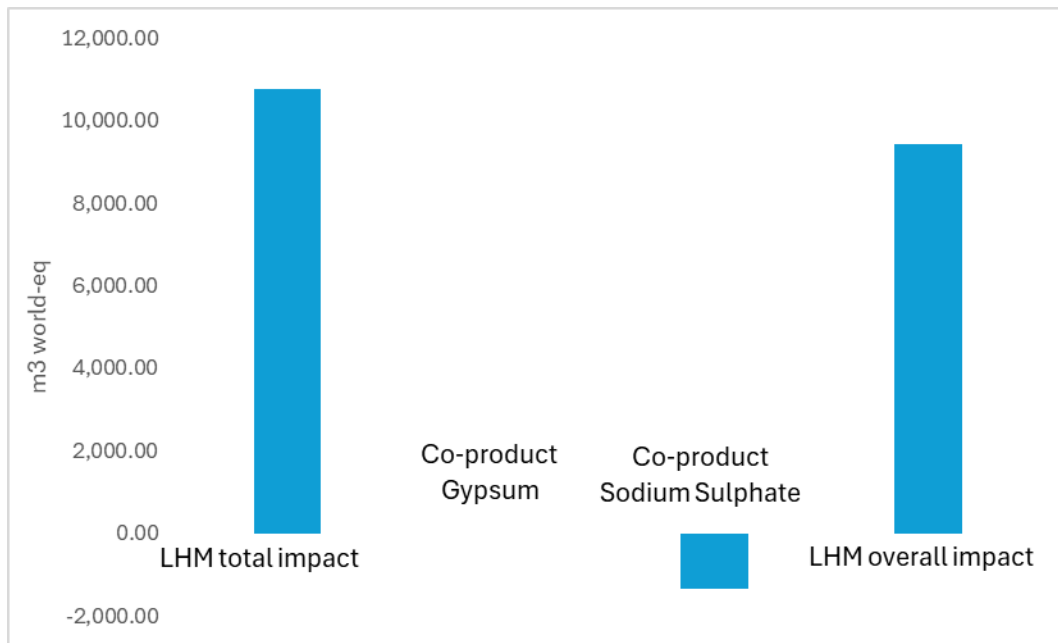


Figure 9: Water scarcity impact of LHM production with co-product offset credits

Due to the processing of ore in the concentrator and flotation, the impact of water consumption is also a key concern that must be assessed for LHM production. As shown in Fig. 9, LHM production exhibits a significant water scarcity footprint, with a total impact of 10,772.98 m³ world-eq per tonne of LHM produced. Co-product modeling through system expansion reduced the overall burden by crediting the displacement of produced sodium sulfate, resulting in a net reduction of approximately 1,500 m³ of water scarcity. As with climate change impacts, gypsum contributed negligibly to water scarcity credits, reinforcing its limited substitution potential or market relevance in the current system boundaries.

Accounting for co-product credits reduced the overall water scarcity impact to 9,422.31 m³ world-eq. Although the impact has decreased, it remains significant, highlighting that water consumption and scarcity are critical environmental concerns in LHM production, especially in regions with stressed freshwater resources or extensive groundwater use during processing.

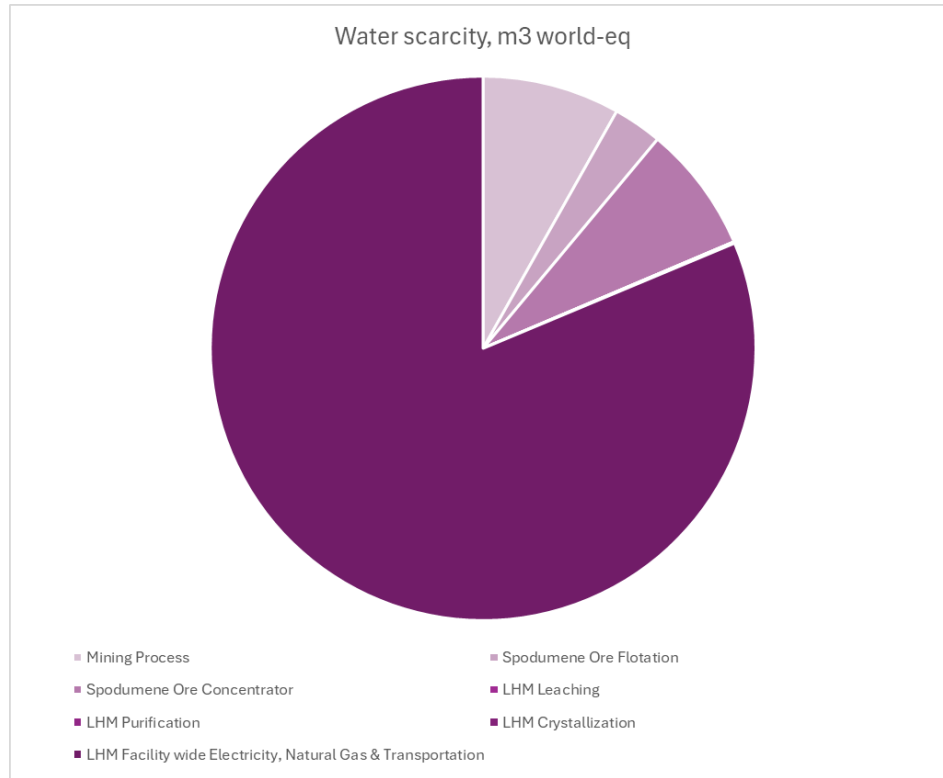


Figure 10: Breakdown of water scarcity impacts across individual unit processes in LHM production, identifying the primary contributors to water consumption

Fig. 10 indicates the highest contribution to water scarcity after offset credits from co-products. The dominant contributor to the total impact was the “LHM Facility-wide Electricity, Natural Gas & Transportation” unit, which alone accounted for 8,760.26 m³ world-eq, or approximately 81.3% of the total water scarcity impact. This category encompasses the background burden of electricity generation, natural gas combustion, and regional/international transportation, each of which has high virtual water use, particularly when assessed using globalized market datasets.

Another key contributor is the process of mining, ore flotation, and concentration, which is evident due to the need for water for ore handling, flotation, and dust suppression. Electricity consumption in crushing and material handling also influences it. In contrast, downstream hydrometallurgical stages such as leaching (0.10 m³ world-eq), purification (0.00 m³ world-eq), and crystallization (9.26 m³ world-eq) contributed negligibly to the total water scarcity impact. Although these steps use chemicals and operate reactors, their marginal contribution indicates that either they withdraw minimal direct water, or their input supply chains are not water-intensive compared to energy processes.

4.4.3. Comparative analysis

A comparative analysis between the present study (2025) and the earlier work by Chordia et al. (2022) [8] reveals several key differences in material inputs, process modeling, market assumptions, and co-product treatment for LHM production in Quebec (refer to Table 4). These differences significantly influence the overall climate change impacts reported: Chordia et al.

estimated a GWP of 9.66 tonnes CO₂-eq, whereas the present study reports a lower impact of 5.46 tonnes CO₂-eq per tonne of LHM.

Table 4: Comparison of current research (2025) versus the Chordia et al. (2022) for provider and category for a Québec LHM mine/concentrator and process plant

Chordia et al (2022)			Current Research (2025)		
Provider material input	Provider inecoinvent	Category	Provider material input	Provider inecoinvent	Category
Mining/Concentrator					
Mining activity input					
Spodumene ore	-	Resource in ground	-	-	-
Blasting	Market for blasting blasting Cutoff, U	GLO	-	-	-
Grinding media	Market for steel, low-alloyed market for forging, steel	GLO	-	-	-
	-	-	Spodumene ore_concentrator	Market for rock crushing rock crushing Cutoff, U	GLO
	-	-	LHM_Spodumene ore_Flotation	-	-
Chemical and reagent input					
Sodium hydroxide	Market for NaOH, without water, in 50% solution state	GLO	Neutralizing agent, sodium hydroxide	Sodium hydroxide to generic market for neutralizing agent neutralizing agent sodium hydroxide equivalent Cutoff, S	GLO
Energy input					
Electricity	Market for electricity, medium voltage	CA-QC	Electricity	Market for electricity, high voltage Cutoff, S	CA-QC
Diesel, mining	Market for diesel, burned in agricultural machinery	GLO	LHM_Mining_Processes_at_Mine	Market for diesel, burned in agricultural machinery Cutoff, S	GLO

Freshwater	Water, lake, CA	Resource in water	Tap water	Market for tap water tap water Cutoff, S	CA-QC
Tailing	Market for non-sulfidic tailing, off-site	GLO	-	-	-
Water, sedimentation pit	Water, lake, CA	Resource in water	Tap water	Market for tap water Cutoff, S	CA-QC

Transportation

-	-	-	(Truck) transport, freight, conveyor belt	Market for transport freight, conveyor belt transport freight. conveyor belt, Cutoff U	GLO
-	-	-	(Rail) transport, freight train	Market for transport freight train transport freight train, Cutoff S	RoW

Process Plant

Chemical and reagent input

Sulfuric acid (H ₂ SO ₄)	Market for sulfuric acid	RoW	Sulfuric acid	Market for sulfuric acid sulfuric acid Cutoff U	RoW
Calcium hydroxide CA(OH) ₂	Market for quicklime, milled packed	RoW	-	-	-
Sodium hydroxide (NaOH)	Market for NaOH, without water, in 50% solution state	GLO	Neutralizing agent, sodium hydroxide	Sodium hydroxide to generic market for neutralizing agent neutralizing agent sodium hydroxide equivalent Cutoff, S	GLO

Energy input

Electricity	Market for electricity, medium voltage	CA-QC	Electricity	Market for electricity, high voltage Cutoff_S	CA-QC
Heat (natural gas)	Market for heat, district or industrial, natural gas	CA-QC	Natural gas, low pressure	Market for natural gas low pressure natural gas low pressure Cutoff, S	RoW

Waste (by-products)

Aluminum silicate (Al ₂ SiO ₅)	Market for waste foundry sand	GLO	Sodium silicate, solid	Market for sodium silicate solid sodium silicate, solid Cutoff U	RoW
Gypsum (CaSO ₄ .2H ₂ O)	Market for waste gypsum	RoW	Gypsum fiberboard	Market for gypsum fiberboard production gypsum production } Cutoff, S	RoW
Sodium sulfate (Na ₂ SO ₄)	-	-	Sodium sulfate, anhydrite	Market for sodium sulfate anhydrite sodium sulfate anhydrite Cutoff, S	RoW

In the mining and concentration phase, the current study replaces blasting and grinding media with rock crushing due to data limitations on blasting processes. Additionally, the model uses sodium hydroxide as a neutralizing agent and shifts energy inputs from medium-voltage electricity (as used by Chordia et al.) to high-voltage electricity and Cutoff-S diesel. Water sourcing also differs: this study substitutes lake water with market tap water and explicitly models transportation via conveyor belts and rail freight, which the 2022 study did not include.

In the process plant phase, both studies include sulfuric acid, but the current study omits calcium hydroxide as a neutralizing agent and updates the classification of sodium hydroxide accordingly. Energy sources also differ, shifting from district/industrial natural gas in Chordia et al. to low-pressure natural gas in the present study.

Key distinctions also emerge in by-product and waste modeling. While the 2022 study credited aluminum silicate derived from waste foundry sand, the present study explored using sodium silicate but rejected it due to excessively high CO₂ credits, which distorted the results. Gypsum, modeled as waste gypsum in the other study, is treated as mineral gypsum production in this study. Importantly, sodium sulfate anhydrite is explicitly modeled as a co-product in the current research, allowing for more accurate credit through system expansion.

These refinements enhance the specificity and transparency of the LCA model, contributing to more robust and contextually relevant environmental impact assessments by adjusting system boundaries, input classifications, and co-product allocation strategies.

4.4.4. Interpretation of Sankey diagram

The Sankey diagram (Fig. 11) generated using OpenLCA provides a visual representation of GHG emissions associated with the cradle-to-gate production of 1 tonne of LHM in Québec. It shows the twenty-five most emission-intensive processes in the defined system boundary, from spodumene ore extraction and concentration at the mine to hydrometallurgical operations, namely leaching, purification, and crystallization, at the LHM production plant. The arrows in the diagram illustrate the flow of CO₂-eq emissions through these processes, with wider arrows indicating greater environmental impact. Emissions are categorized into two types: direct emissions, occurring on-site during specific processes (e.g., fuel combustion in kilns or dryers), and upstream

emissions, generated in the supply chain before the process (e.g., from producing or transporting input materials and energy).

This diagram shows that most emissions occur upstream rather than on-site at the LHM production facility. The defined unit processes, such as the production of 1 ton of LHM, crystallization, and leaching, involve upstream CO₂ emissions, with the "LHM Facility Wide" process exhibiting upstream emissions totaling 5.13 tonnes CO₂ eq. Of this, approximately 3.099 tonnes CO₂ eq. comes from upstream emissions related to the combustion of natural gas used across the facility for boilers, the calciner, the aluminium silicate dryer, and other thermal units. Transport of the spodumene concentrate from the mine to the plant also contributes to emissions: 0.2912 tonnes CO₂ eq. from train freight and 0.2851 tonnes CO₂ eq. from lorry freight.

Additionally, diesel used for mining equipment (e.g., drillers and blasters) contributes significantly (amounting to 1.08 tonnes CO₂ eq.). In addition, the diagram accounts for emissions from co-products (shown in green), which can potentially offset carbon. Anhydrous sodium sulfate, which is co-produced during the crystallization process, results in a carbon offset of approximately 1.008 tonnes CO₂ equivalent. This conclusion is consistent with Figures 7 and 8.

Sankey diagrams in LCA do not depict cumulative or additive emissions across the entire system boundary. Instead, each node in the Sankey diagram (e.g., "Purification" or "LHM Facility Wide") shows the direct emissions from that process as well as the upstream emissions required to support it. However, because upstream processes (such as 'market for heat' or 'market for transport') serve multiple downstream nodes, their emissions appear multiple times, once per connection. Therefore, while Sankey diagrams effectively visualize hotspots and process-level dependencies, they should not be used to determine total emissions.

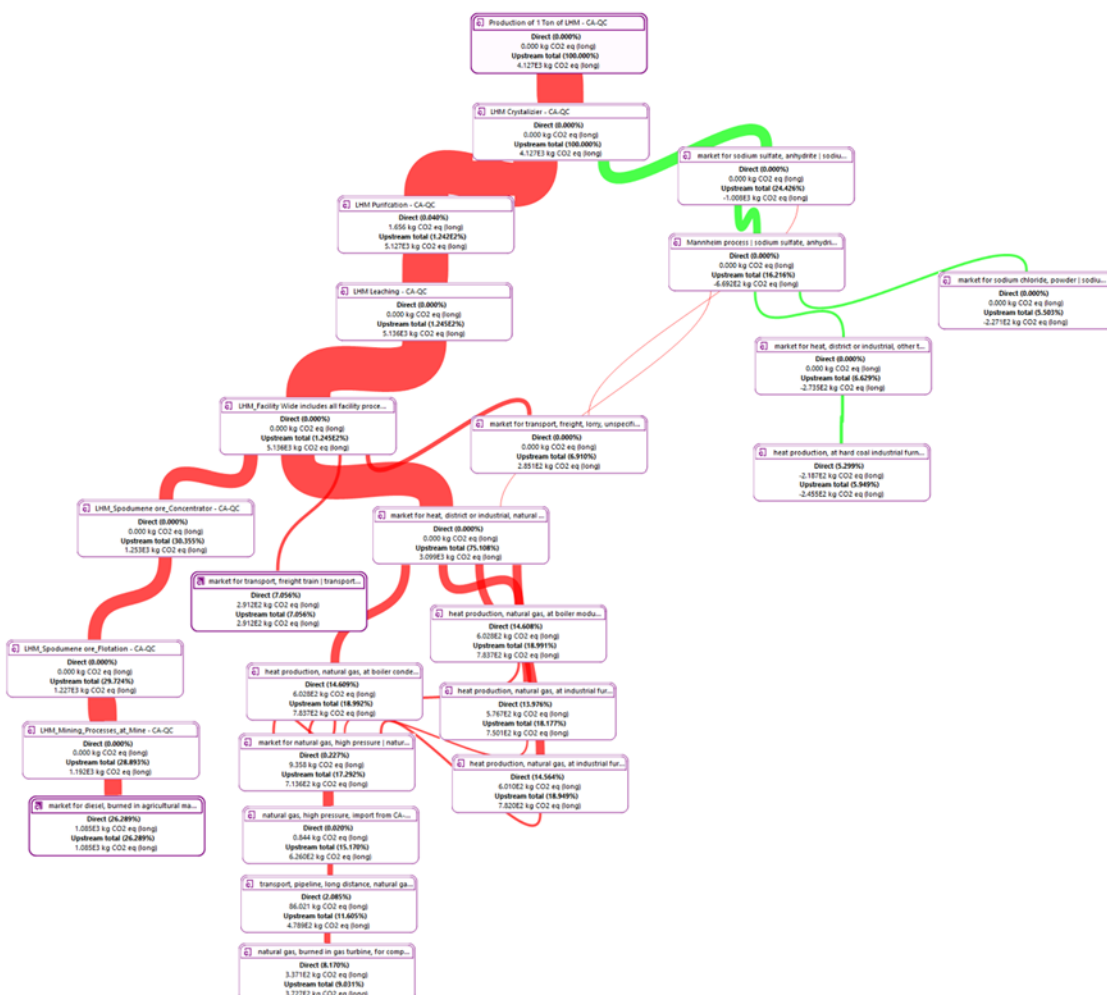


Figure 11: Sankey diagram to produce one ton of LHM in Québec. The red line represents CO₂ emissions and green lines represents CO₂ credits from co-products

4.5. Concluding remarks and perspectives

The main environmental impacts of producing one tonne of LHM arise from energy consumption (including electricity, diesel, gasoline, and natural gas), water usage, waste generation, and by-products. Per 1,000 kg of LHM, short-term climate change totaled 4,395.42 kg CO₂ eq, and the water scarcity impact reached 9,422.31 m³ after accounting for the sodium sulfate by-product. Assessment covered a mine and processing plant in Québec, Canada. The cradle-to-gate LCA covers the process from mining to battery-grade material. Impacts were calculated using *OpenLCA v2.3*, *Ecoinvent v3.8* database, and the *Impact World+ Midpoint v2.0.1* method, based on data from a spodumene mine and LHM production plant in Québec, Canada. Natural gas use in the process plant produced the highest CO₂ emissions at the Québec mine and processing facility. This case study has shown that even though Québec has lower GWP linked to renewable hydroelectric power, electrification of the process calciner can help to lower the CO₂ generation at the process plant.

Additionally, electrification of mining equipment (excavators, dump trucks, graders, etc.) and the replacement of fossil fuel vehicles with electric models could eliminate the impact from fuel consumption. This work demonstrates that an LCA can help identify hotspots and

opportunities for improvement of products, herein battery-grade LHM, that are critical to a sustainable future. The province of Québec continues to work towards a “green battery” with zero waste, and performing LCA on critical mineral extraction and processing in the province is a necessary step to provide valuable information and insights for the decision-makers and ensure progress toward this goal.

References

1. G. Finnveden and J. Potting, "Life cycle assessment," in *Encyclopedia of Toxicology*, 3rd ed., Elsevier, 2014, pp. 74–77, doi: 10.1016/B978-0-12-386454-3.00627-8.
2. L. Rigamonti and E. Mancini, "Life cycle assessment and circularity indicators," *The International Journal of Life Cycle Assessment*, vol. 26, no. 10, pp. 1937–1942, Sep. 2021, doi: 10.1007/s11367-021-01966-2.
3. J. Porzio and C. D. Scown, "Life-Cycle Assessment Considerations for Batteries and Battery Materials," *Advanced Energy Materials* 2021, Vol. 11, Page 2100771, Jul. 2022, doi: 10.1002/aenm.202100771.
4. European Commission Joint Research Centre, "Life Cycle Assessment," European Platform on LCA, accessed Apr. 9, 2025. [Online]. Available: <https://eplca.jrc.ec.europa.eu/lifecycleassessment.html>.
5. NAATBatt International, "The Conundrum of the North American Lithium-Ion Battery Supply Chain," NAATBatt International, accessible online, Accessed Apr. 9, 2025, Available: <https://naatbatt.org/the-conundrum-of-the-north-american-lithium-ion-battery-supply-chain/>.
6. BloombergNEF, "China Drops to Second in BloombergNEF's Global Lithium-Ion Battery Supply Chain Ranking as Canada Comes Out on Top," BloombergNEF, 2024, Feb. 2024, Accessed: Apr. 10, 2025 [Online]. Available: <https://about.bnef.com/insights/commodities/china-drops-to-second-in-bloombergnefs-global-lithium-ion-battery-supply-chain-ranking-as-canada-comes-out-on-top/>.
7. Government of Canada – Natural Resources Canada, "Critical minerals: an opportunity for Canada," Canadian Critical Minerals Campaign, [Online]. Available: <https://www.canada.ca/en/campaign/critical-minerals-in-canada/critical-minerals-an-opportunity-for-canada.html>. Accessed: Apr. 10, 2025.
8. M. Chordia, S. Wickerts, A. Nordelöf, and R. Arvidsson, "Life cycle environmental impacts of current and future battery-grade lithium supply from brine and spodumene," *Resources, Conservation and Recycling* 2022, Vol. 187, Page 106634, Dec. 2022, doi: 10.1016/j.resconrec.2022.106634.
9. A. Mas-Fons, R. Arduin, P. Loubet, T. Pereira, A. Parvez, and G. Sonnemann, "Carbon and water footprint of battery-grade lithium from brine and spodumene: A simulation-based LCA," *Journal of Cleaner Production* 2024, Vol. XX, Page YYYYY, 2024, doi: 10.1016/j.jclepro.2024.YYYYY.
10. S. Ibarra-Gutiérrez, J. Bouchard, M. Laflamme, and K. Fytas, "Assessing the potential of Quebec lithium industry: Mineral reserves, lithium-ion batteries production and greenhouse gas emissions," *Resources Policy* 2021, Vol. 74, Page 102371, Dec. 2021, doi: 10.1016/j.resourpol.2021.102371.
11. Met-Chem, "NI 43-101 Technical Report: Feasibility Study Update on the Whabouchi Lithium Deposit and Hydromet Plant (Revised)," Met-Chem, May 31, 2019, [Online]. Available: <https://minedocs.com/22/Whabouchi-TR-05312019.pdf>. Accessed: Jul. 24, 2025.
12. Livent Corporation, "Pre-Feasibility Study (PFS) on the Whabouchi Lithium Project," Livent Corporation, Dec. 31, 2022, [Online]. Available: <https://minedocs.com/25/Whabouchi-PFS-12312022.pdf>. Accessed: Feb. 23, 2025.
13. Nunavik Lithium Inc., "Environmental and Social Overview: Executive Summary," Nunavik Lithium Inc., 2025, [Online]. Available: <https://ehq-production-canada.s3.ca->

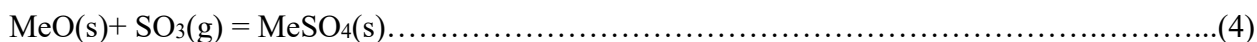
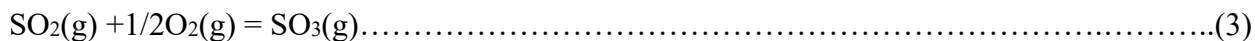
- central-
 1.amazonaws.com/5290e583a31cf7ed8fd6679314dcc1e4d5508276/original/1702317923/a87adbdc5a24aac86c7989ffead2805_NLI_EES_R%C3%A9sum%C3%A9Ex%C3%A9cutif.pdf. Accessed: Feb. 23, 2025.
14. Nunavik Lithium Inc., “Environmental and Social Overview, Volume 1: Chapters 1 to 6,” Nunavik Lithium Inc., Oct. 2023, [Online]. Available: https://ehq-production-canada.s3-central-1.amazonaws.com/e598bbf355ce72918a127ab76d9d085010fdda88/original/1702310809/dff812165e5d00c0f6f993f52f7e7ca1__NLI_EES_Vol._1_Chap._1_%C3%A0_6_%C3%A9d_octobre_2023.pdf. Accessed: Feb. 23, 2025.
 15. Nunavik Lithium Inc., “Environmental and Social Overview, Volume 2: Chapters 7 to 15,” Nunavik Lithium Inc., Oct. 2023, [Online]. Available: https://ehq-production-canada.s3-central-1.amazonaws.com/edecfdb6127fe79ab443d716777f01280ed2893b/original/1702310896/107e36b42ce17c1e1a5bf5225d54067a__NLI_EES_Vol._2_Chap._7_%C3%A0_15_%C3%A9d_octobre_2023.pdf. Accessed: Feb. 23, 2025.
 16. C. Bulle, M. Margni, L. Patouillard, A.-M. Boulay, G. Bourgault, V. De Bruille, V. Cao, M. Hauschild, A. Henderson, S. Humbert, S. Kashef-Haghighi, A. Kounina, A. Laurent, A. Levasseur, G. Liard, R. K. Rosenbaum, and P.-O. Roy, “IMPACT World+: a globally regionalized life cycle impact assessment method,” *International Journal of Life Cycle Assessment*, vol. 24, no. 9, pp. 1653–1674, 2019, doi: 10.1007/s11367-019-01583-0.
 17. International Energy Agency (IEA), “Methane and Climate Change,” *Methane Tracker 2021*, IEA, Paris, 2021. [Online]. Available: <https://www.iea.org/reports/methane-tracker-2021/methane-and-climate-change>. Accessed: Jun. 10, 2025.

Chapter 5 - Optimization of nickel extraction from nickel sulfide ore

5.1. Background

The prolonged exploitation of nickel sulfide deposits has significantly depleted reserves of high-grade ores, shifting industrial and research interest toward the processing of low-grade nickel sulfide resources. These low-grade ores typically contain more than 90% gangue minerals, which substantially increases the complexity and cost of processing [1]. As discussed in Section 3.3.2.1, industrial operations predominantly employ smelting for metal recovery; however, this method is associated with considerable metal losses, up to 70% cobalt, and notable amounts of nickel and copper are retained in the slag, while achieving relatively low recovery efficiencies [2]. Given the strategic importance of nickel and cobalt in manufacturing electrode active materials for LIBs, it is critical to develop extraction processes that maximize recovery while minimizing waste to ensure the sustainable availability of these metals.

For efficient extraction from low-grade nickel sulfide ores, researchers have increasingly investigated integrated pyrometallurgical–hydrometallurgical processes, with the roasting–leaching method emerging as a promising alternative to conventional smelting. This method not only offers higher recovery potential but is also more environmentally compatible. Roasting involves the thermal conversion of mineral sulfides into oxides, sulfates, or chlorides [3]. In nickel processing, the two predominant roasting approaches are chlorination and sulfation roasting. Chlorination roasting uses chlorine-containing agents, such as chlorine gas, hydrogen chloride, calcium chloride, sodium chloride, magnesium chloride, or aluminium chloride, to convert the target metals into soluble chlorides. Although this method achieves high reactivity and extraction rates, the corrosive nature of chlorination severely impacts equipment longevity and increases operational costs. In contrast, sulfation roasting oxidizes sulfides in air or oxygen-enriched atmospheres to form water-soluble sulfates. This technique offers high efficiency, compact processing, and adaptability to a variety of feedstocks, making it a preferred option for low-grade nickel ores [4]. The principal reactions occurring during sulfation roasting are as follows (1–4):



Here, Me denotes Ni, Co, Cu, or Fe. Reaction (1) represents direct sulfation, whereas reactions (2)–(4) constitute the indirect sulfation pathway, which is generally dominant during the process. To improve sulfation efficiency, oxidizing salt additives are frequently employed as catalysts.

Sodium sulfate (Na_2SO_4) has been widely recognized for its ability to enhance nickel extraction due to its catalytic action on sulfide minerals. Its relatively low cost, averaging around US\$0.37 per kg, also supports its industrial viability [6]. Li et al. investigated the mechanism of sodium sulfate addition and proposed a two-stage process: heating and holding. During heating, a liquid-phase double salt ($\text{Na}_2\text{Me}(\text{SO}_4)_2$) forms, which dissolves the excess SO_3 produced during sulfide oxidation and prevents oxygen from converting the NiS core into NiO or NiFe_2O_4 . In the holding stage, NiS gradually transforms into NiSO_4 or $\text{Na}_2\text{Ni}(\text{SO}_4)_2$ [5]. Similarly, Sun et al. applied sodium sulfate for the simultaneous recovery of Ni, Cu, and Co from low-nickel matte

(25.21 wt% Ni), achieving recovery rates of 95%, 99%, and 94%, respectively. Their mechanistic study confirmed Li et al.'s findings and further identified the formation of eutectic compound sulfates $[\text{MeSO}_4 \cdot \text{Na}_2\text{SO}_4]$ (Me = Ni, Cu, Co), which enhance leaching by facilitating oxygen diffusion while suppressing the escape of SO_2/SO_3 gases, thereby maximizing sulfation efficiency [2]. In addition to sodium sulfate, other additives such as ammonium sulfate have been explored, often in combination with sodium sulfate [8,9].

In this study, a novel dual salt additive system comprising sodium sulfate and potassium sulfate is investigated to optimize roasting parameters for enhanced nickel recovery. The ore sample was subjected only to crushing and grinding, without flotation, resulting in a feed material with a high impurity content that increases the complexity of the extraction process. As outlined in Chapter 4, sodium sulfate is generated as a co-product during lithium hydroxide production in Quebec, Canada; its application in the present work therefore aligns with circular economy principles by enabling its direct reuse within the Canadian market. Although researchers have examined potassium sulfate for laterite (oxide) ores, its effect on nickel sulfide ores remains unexplored. This investigation, therefore, addresses both the technical challenge of processing high-impurity nickel sulfide ores and the strategic need for sustainable resource utilization, while introducing a new salt additive combination to improve metal recovery efficiency.

5.2. Experiments

5.2.1. Materials and Reagents

This study obtained nickel sulfide ore from the Glencore Raglan Mine, Canada. The ore had been pre-crushed and reduced to powder form by Glencore prior to delivery. Sigma Aldrich supplied sodium sulfate ($\geq 99\%$, anhydrous) and potassium sulfate ($\geq 99\%$) as roasting additives. Deionized water for leaching experiments came from the laboratory's Milli-Q water system.

The chemical composition of the ore is presented in Table 5, showing that nickel content is only 5.093 wt.%. Iron and magnesium are the dominant elements, with concentrations of 29.61 wt.% and 14.71 wt.%, respectively. To identify the major mineral phases, X-ray diffraction (PANalytical Empyrean) analysis was conducted. The resulting XRD pattern (Fig. 12) indicates the presence of pyrrhotite ($\text{Fe}_{0.87}\text{Ni}_{0.02}\text{S}$), pentlandite ($\text{Fe}_{4.8}\text{Ni}_{4.2}\text{S}_8$), lizardite ($\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$), and chalcopyrite (CuFeS_2). Pentlandite is identified as the primary nickel-bearing mineral, while pyrrhotite and lizardite constitute the major gangue phases.

Table 5: Chemical composition of nickel sulfide ore

Ni	Cu	Co	Fe	Mg	S	Si	Al	Ca	Cr	Mn	Ti	K
5.093	1.863	0.0879	29.61	14.71	19.03	8.7	1.07	1.84	0.162	0.078	0.075	0.02

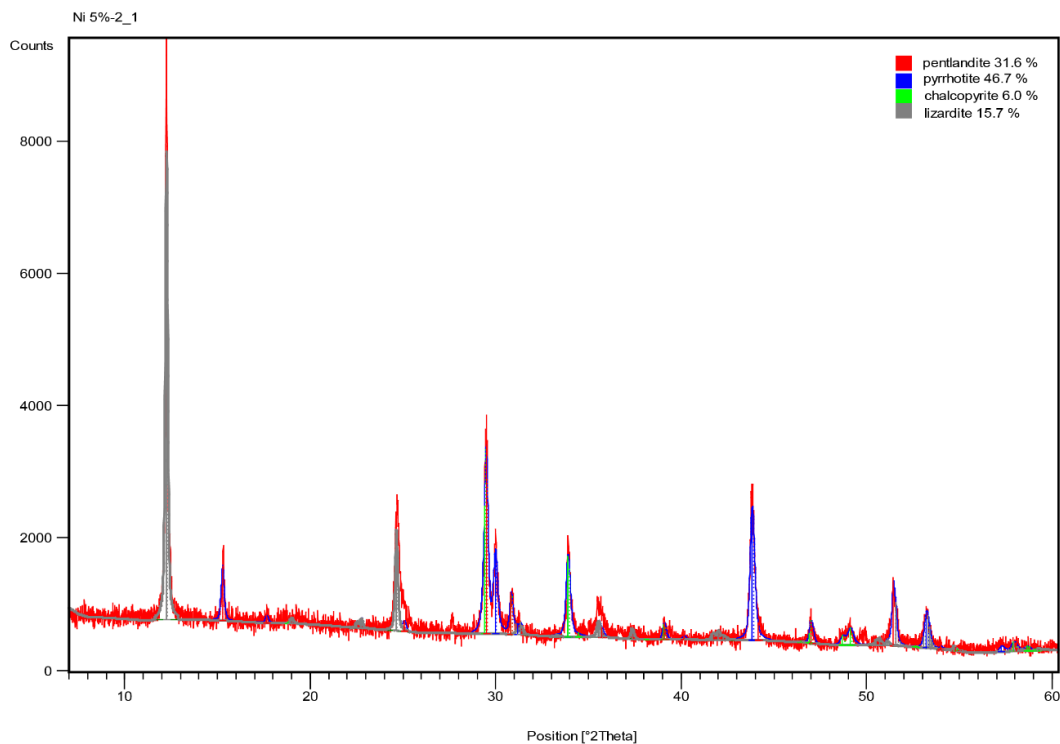


Figure 12: XRD of nickel sulfide ore

A scanning electron microscope (SEM, Hitachi Regulus 8230) coupled with energy-dispersive X-ray spectroscopy (EDS) characterized the particle morphology and surface composition of the ore. SEM imaging (Fig. 13a) shows that the maximum particle size is 30.2 μm . Higher magnification images reveal that the particles are irregular in shape, with fine particles loosely agglomerated onto larger grains. In backscattered electron (BSE) (Fig. 13b) images, brighter regions are attributed to Ni-rich pentlandite, while darker regions correspond to gangue minerals such as lizardite. Elemental mapping by EDS (Fig. 14) confirms the distribution of pentlandite (a, b, d) and chalcopyrite (a, c, d), as well as the gangue phase lizardite (e, f, g).

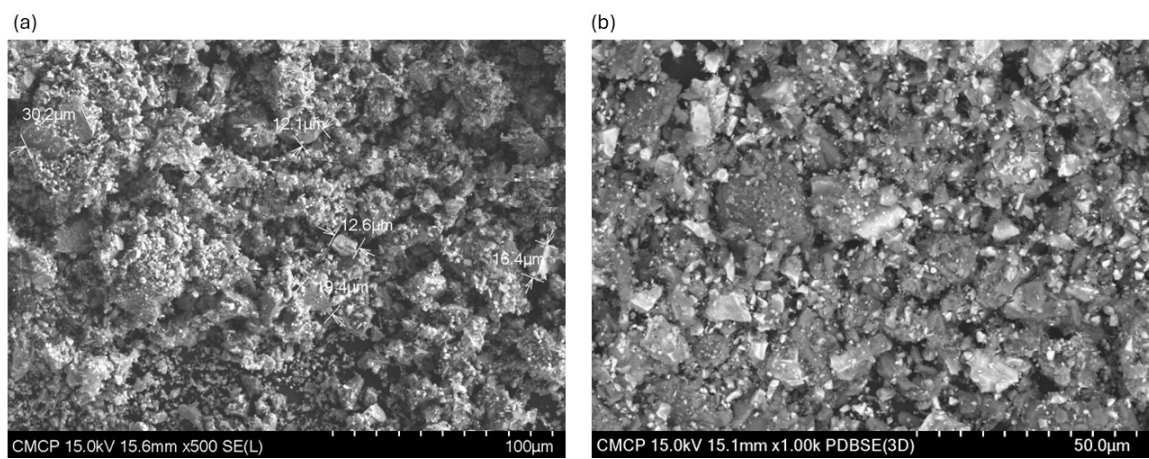


Figure 13: SEM images of nickel ore

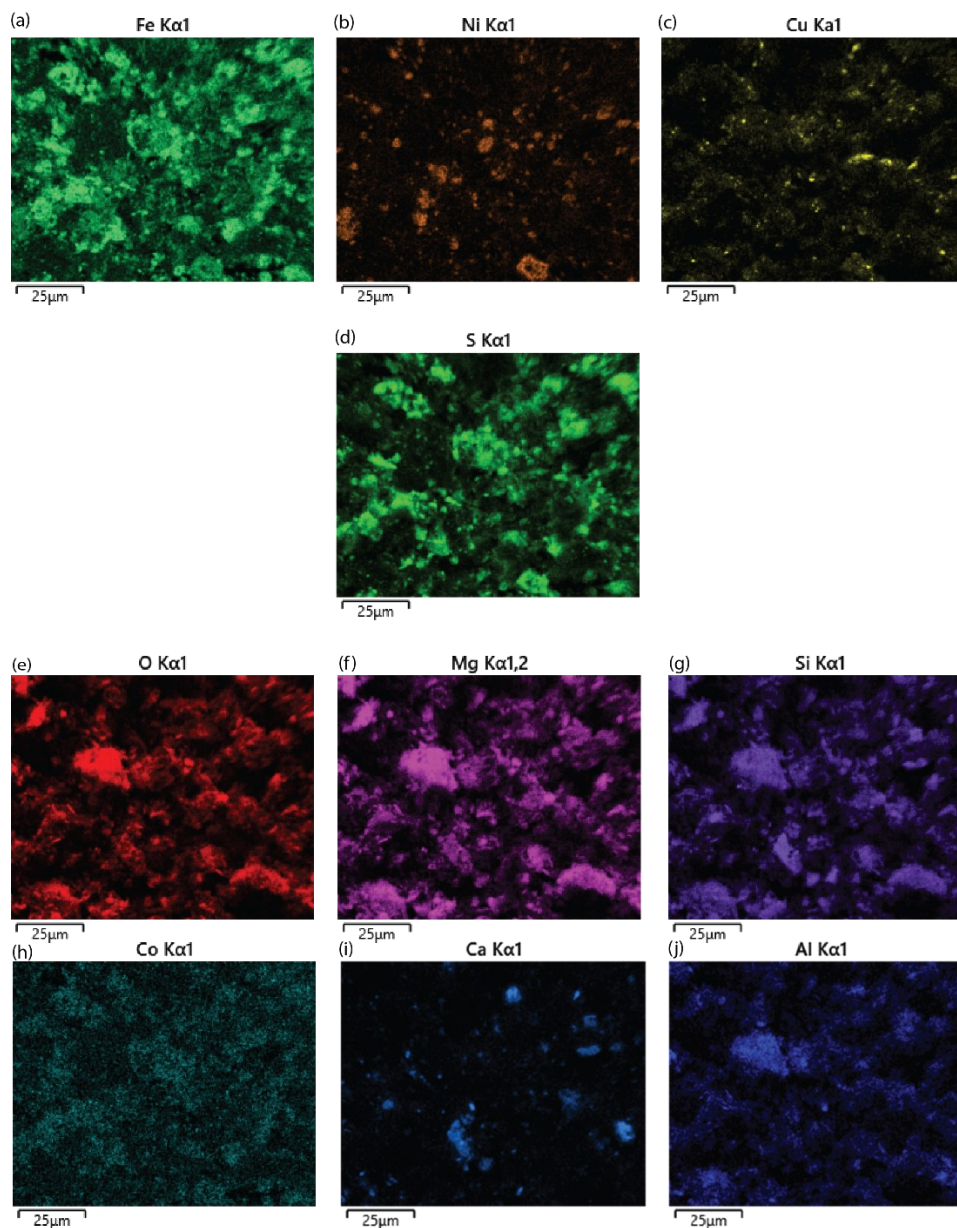


Figure 14: EDS mapping of nickel sulfide ore indicating the different mineral phases present in the ore

5.2.2. Methods

The complete roasting–leaching process is illustrated in Fig. 15. Four roasting parameters were optimized in this study: roasting temperature, roasting time, Na_2SO_4 dosage, and K_2SO_4 dosage.

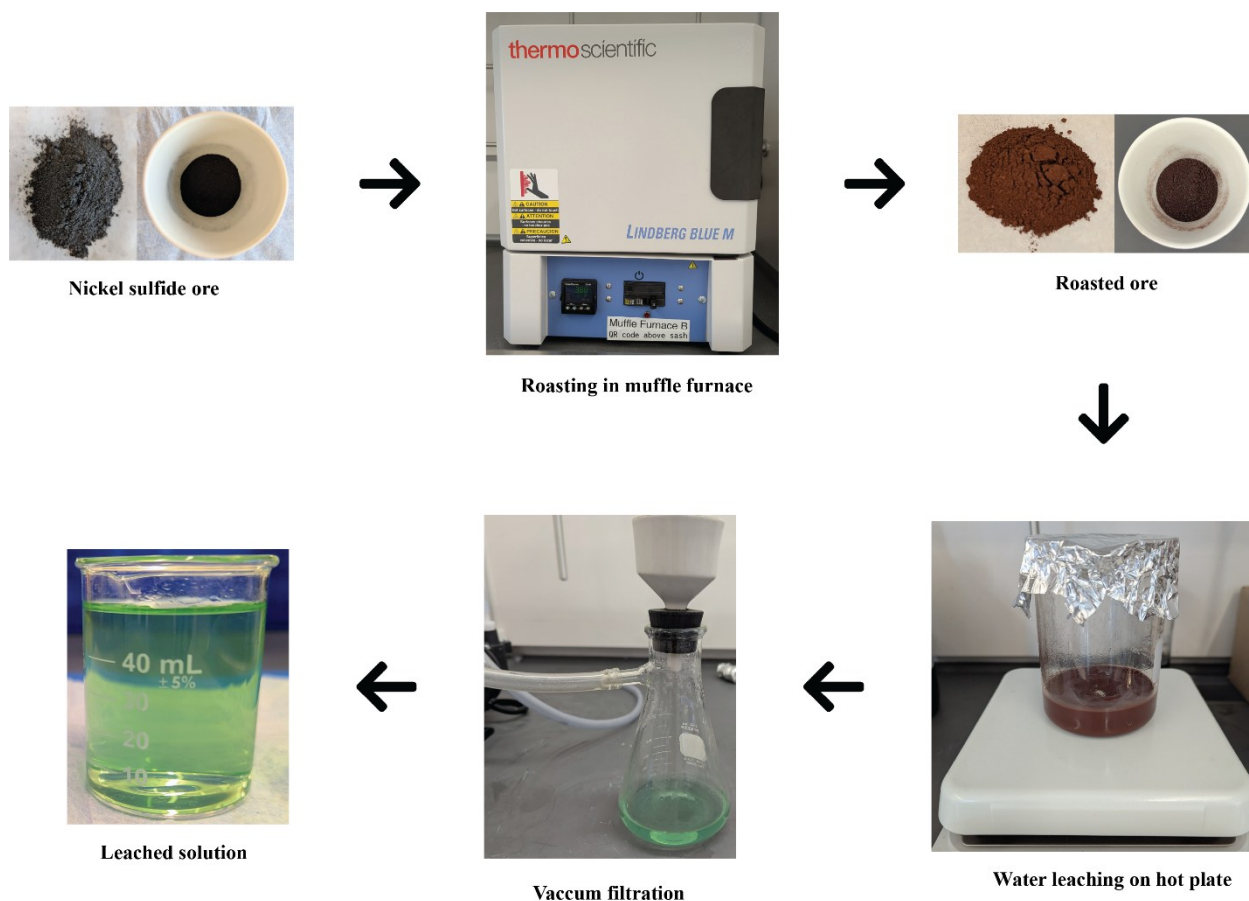


Figure 15: The sulfation roasting-leaching process flowchart employed for the nickel recovery

For each experiment, 10 g of ore was thoroughly mixed with the specified amount of salt additive and placed in a high-alumina crucible. The crucible was then inserted into a muffle furnace for the designated duration, with the furnace outlet vent left partially open to allow the escape of flue gases. The parameters, such as temperature, time, and salt dosages, were varied according to the ranges listed in Table 6. The cell highlighted in gray were the values kept constant during the optimization of particular parameter. Upon completion of roasting, the furnace was allowed to cool naturally to room temperature, after which the roasted clinker was collected for leaching.

Table 6: Roasting conditions for optimization of each parameter

	Optimizing parameters			
	Temperature (°C)	Time (hrs)	Na ₂ SO ₄ (g)	K ₂ SO ₄ (g)
Temperature (°C)	450 to 700	650	650	650
Time (hrs)	3	1 to 3.5	2.5	2.5
Na ₂ SO ₄ (g)	5	5	1 to 6	4
K ₂ SO ₄ (g)	0	0	0	0.25 to 1.75
Furnace heating rate (°C/min)	5	5	5	5

Leaching conditions were kept constant for all roasting parameter tests. Water leaching was carried out using deionized water on a hot-stir plate at 80 °C, with a stirring speed of 500 rpm and a liquid-to-solid ratio of 5:1 (mL:g). After leaching, the slurry was vacuum-filtered to separate the leachate from the solid residue. The concentrations of metal ions in the leachate were measured using inductively coupled plasma mass spectrometry (ICP-MS, Agilent Technologies, 7700 Series), and the extraction efficiency (α_i) was calculated according to Eq. (5):

$$\alpha_i = \frac{C_i v_i}{w_i m} \times 100\%$$

Where α_i (%) is the conversion of the metal elements, C_i (g/L) is the concentration of metal ions in the mixed solution of the leaching solution and the washing solution, v_i (L) is the volume of the leaching solution, w_i (wt.%) is the content of metal elements in the ore, and m (g) is the mass of the ore powders used in the roasting procedure [7]. Each experiment was performed in duplicate, and the average values were reported.

Following optimization of all four roasting parameters, the solid residues were washed three times with deionized water, dried in an oven at 100 °C for 1 h, and subjected to further characterization. XRD analysis was performed to examine the crystalline structure of roasted clinkers obtained using different potassium sulfate dosages. SEM-EDS analysis was carried out on roasted products prepared under optimized temperature and time conditions for three cases: (i) no additive, (ii) sodium sulfate additives, and (iii) dual sulfate additives.

5.3. Results and Discussion

Effect of roasting temperature

The effect of roasting temperature on the extraction of Ni using the sulfation roasting–water leaching technique was investigated first. As summarized in Table 6, roasting was conducted for 3 h at a heating rate of 5 °C/min with 5 g of Na₂SO₄. According to the predominance diagram of Ni_{4.5}Fe_{4.5}S₈–CuFeS₂–SO₂–O₂ [5], the temperature range that favors enhanced Ni extraction is between 500 °C and 800 °C. Based on this information and previous studies, the temperature range selected for this study was 450–700 °C.

As shown in Fig. 16, Ni extraction increases with increasing temperature, reaching a maximum before decreasing, in agreement with earlier reports [2,8]. Between 450 °C and 600 °C, the increase in Ni extraction is steep, varying from 16% to 54% respectively. The maximum extraction efficiency of 59% is achieved at 650 °C, after which it decreases to 55% at 700 °C. Li et al. reported that heating Ni sulfide ore to 650 °C leads to the gradual conversion of the NiS phase to Na₂Ni(SO₄)₂ (double salt), which suppresses the escape of SO₃ gas and promotes sulfation, thereby achieving high extraction efficiency [5].

In contrast, Fe shows a relatively high extraction efficiency of 17% at 450 °C, which decreases with increasing temperature. This decline can be attributed to the thermal decomposition of FeSO₄ at high temperatures (around 700 °C) into ferric oxide and SO₂ [2], as described by Reaction (6):



Thus, the optimized roasting temperature is 650 °C.

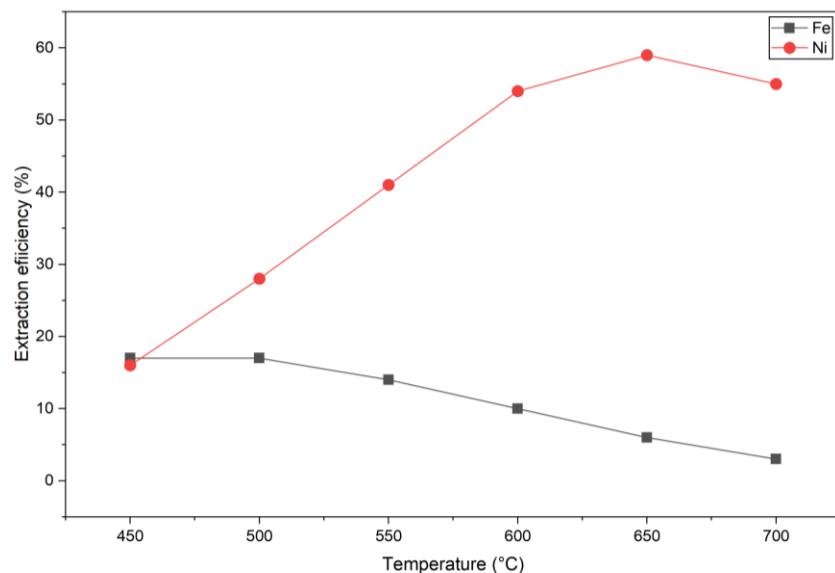


Figure 16: Effect of temperature on Ni and Fe recovery from nickel sulfide ore using only sodium sulfate as additive

Effect of roasting time

The influence of roasting time on Ni extraction was investigated at 650 °C with 5 g of Na₂SO₄ and a heating rate of 5 °C/min, as summarized in Table 6. Roasting durations ranging from 1 h to 3.5 h were evaluated.

As shown in Fig. 17, Ni extraction gradually increases with roasting time, reaching a maximum of 60% at 2.5 h, after which the extraction efficiency plateaus. This trend is consistent with previous studies [9], where the initial rise in extraction is attributed to the increased time available for sulfation of NiS. Beyond 2.5 h, the extraction efficiency shows negligible improvement, indicating that the reaction has essentially reached completion.

Similarly, Fe extraction shows minimal variation with time, plateauing at approximately 6%. Extending the roasting duration beyond 2.5 h offers no significant gain in extraction but results in unnecessary energy consumption. Therefore, the optimum roasting time for maximizing Ni recovery while maintaining process efficiency is 2.5 h.

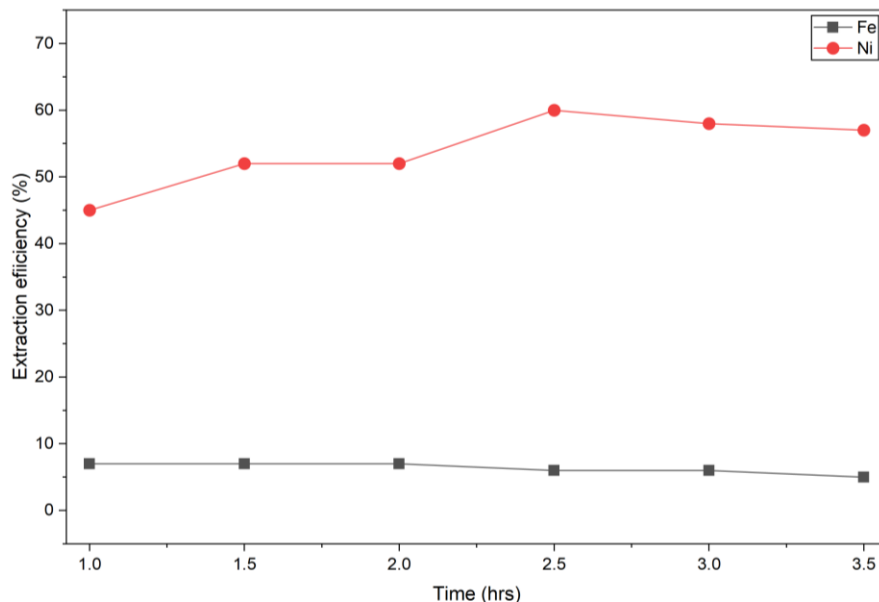


Figure 17: Effect of roasting duration on the Ni and Fe recovery using only sodium sulfate as salt additive

Effect of sodium sulfate dosage

The influence of sodium sulfate (Na_2SO_4) addition on Ni and Fe extraction was investigated at a roasting temperature of $650\text{ }^\circ\text{C}$, a holding time of 2.5 h, and a heating rate of $5\text{ }^\circ\text{C min}^{-1}$. As shown in Fig. 18, Ni extraction efficiency increased from 47% at the lowest dosage (1 g) to a maximum of 63% at a dosage of 4 g. This enhancement aligns with previous studies, which have demonstrated that Na_2SO_4 acts as a sulfation promoter during roasting by facilitating the formation of sodium–metal double sulfates that are more readily leached [9].

A key mechanism proposed in literature is the reaction between Na_2SO_4 and SO_3 (generated during sulfide oxidation) to form sodium pyrosulfate ($\text{Na}_2\text{S}_2\text{O}_7$). This compound exists as a liquid phase at roasting temperatures and can permeate the product layer, increasing contact with unreacted particle surfaces and thus enhancing sulfation completeness [9]. The improved reaction kinetics result in higher Ni conversion to NiSO_4 , explaining the substantial extraction increase observed up to the 4 g dosage, beyond this optimum, further Na_2SO_4 addition slightly reduced Ni extraction.

Fe extraction remained low ($\sim 3\%$) across all Na_2SO_4 dosages, indicating that sodium sulfate selectively enhances Ni sulfation while suppressing Fe dissolution under the tested conditions. Thus, the optimum Na_2SO_4 dosage for maximizing Ni recovery in this system is 4 g.

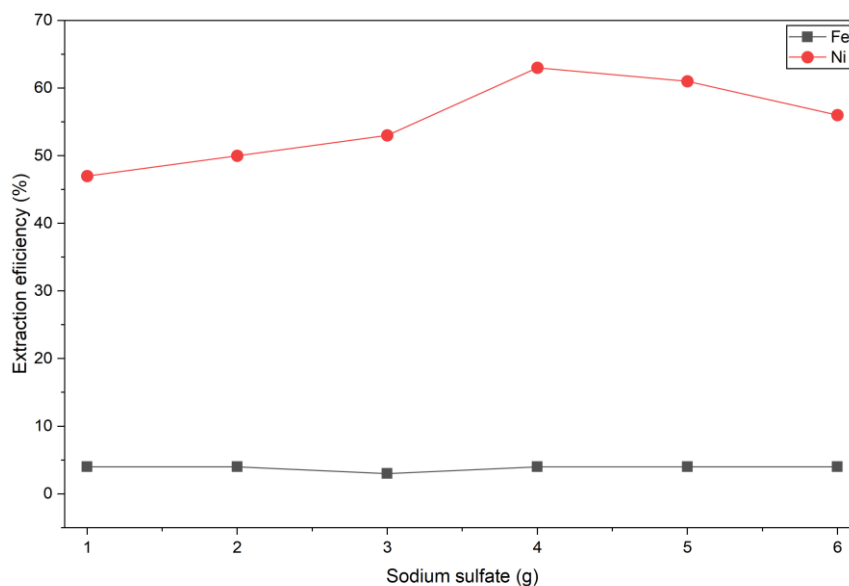


Figure 18: Effect of various sodium sulfate dosage on Ni and Fe recovery

Effect of potassium sulfate

This study investigates the synergistic effect of a novel dual salt additive (potassium sulfate and sodium sulfate). In Fig. 18, sodium sulfate alone has already been shown to enhance Ni extraction, achieving up to 63% under the optimized roasting conditions. To study the effect of potassium sulfate, experiments were conducted at 650 °C for 2.5 h with 4 g of Na_2SO_4 .

As shown in Fig. 19 (a), the addition of potassium sulfate significantly improved the extraction efficiency. Increasing the K_2SO_4 dosage led to higher Ni recovery, reaching 79% at 1.25 g. This enhancement can be attributed to the formation of liquid pyrosulfates, as reported in studies on various alkali salts used for recovering valuable metals from saprolite ores. The liquid pyrosulfates wet the ore and decompose to release SO_3 , which subsequently reacts with unreacted nickel oxides to form nickel sulfates [10]. Increasing the K_2SO_4 dosage beyond 1.25 g may cause incomplete Ni extraction due to the formation of solid Ni/Cu–Co–Fe spinel phases, which hinder nickel liberation and sulfation. XRD analysis (Fig. 19 (b)) of the roasted product confirms the formation of a metal–potassium double salt, $\text{K}_2\text{Ni}_2\text{O}_{12}\text{S}_3$, indicative of enhanced reaction kinetics through pyrosulfate formation [2]. Analysis detected additional phases, including nickel and copper sulfates, and a small amount of NiS, which likely explains the incomplete nickel extraction.

Fe extraction showed only minor variations, remaining approximately 5%. XRD results indicate that most of the iron was retained as oxides or incorporated into Na–K jarosite complexes, consistent with low co-extraction of iron and effective separation from nickel. Therefore, the study selected 1.25 g as the optimized K_2SO_4 salt dosage.

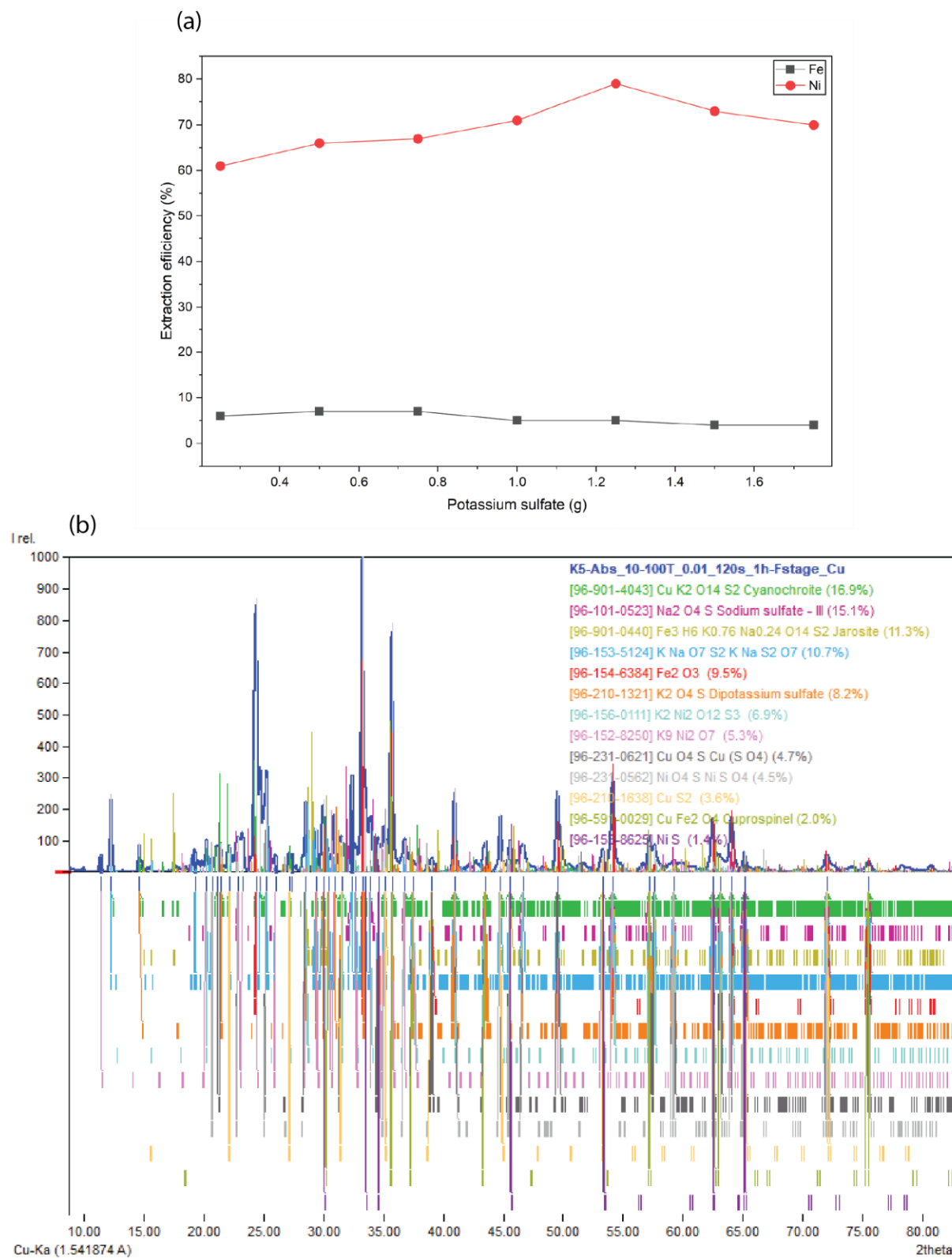


Figure 19: (a) Influence of different potassium sulfate dosages on Ni and Fe recovery in a dual salt additive system (b) XRD pattern of the roasted clinker using the optimized potassium sulfate dosage (1.25 g)

Post-roasting analysis

SEM analysis examined the impact of sodium sulfate and potassium sulfate as additives on roasted products. For comparison, under the optimized roasting conditions (650 °C and 2.5 h), three experiments were analyzed: one without any additives, one with the optimized dosage of sodium sulfate (4 g), and one with the optimized dosage of dual salt, sodium and potassium sulfate (4 g and 1.25 g, respectively).

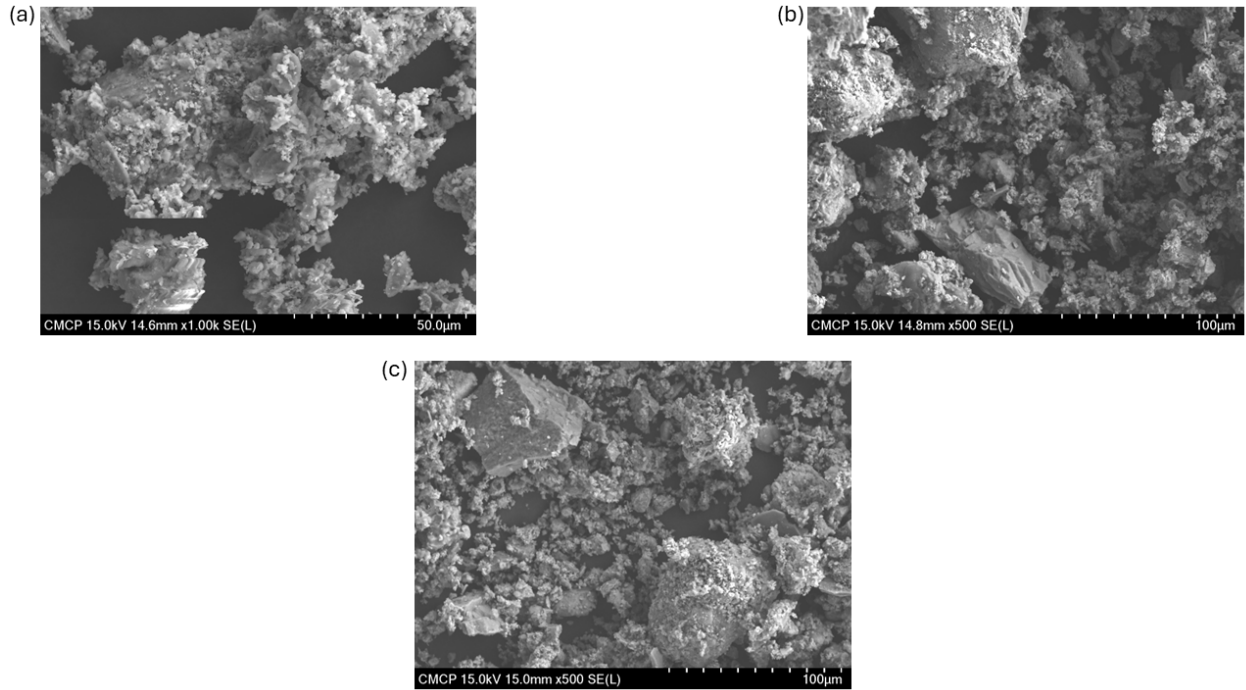


Figure 20: SEM analysis of roasted clinkers (a) without any additives (b) with sodium sulfate (c) with dual salt additives

In Fig. 20(a), the roasted product without additives shows irregular and coarse morphologies with dense structures and limited porosity, indicative of incomplete phase transformation. The particles appear loosely agglomerated, reflecting poor mineral breakdown. From EDS mapping (Fig. 21) and Table S1, the sample shows the presence of impurities in the form of Fe oxides and Mg-silicates, corresponding to the primary gangue mineral lizardite. Ni is mainly associated with Fe and S, consistent with its occurrence in pentlandite. The following

analysis suggests that in the absence of salt additives, Ni largely remains locked within pentlandite, which explains the lower extraction efficiency observed.

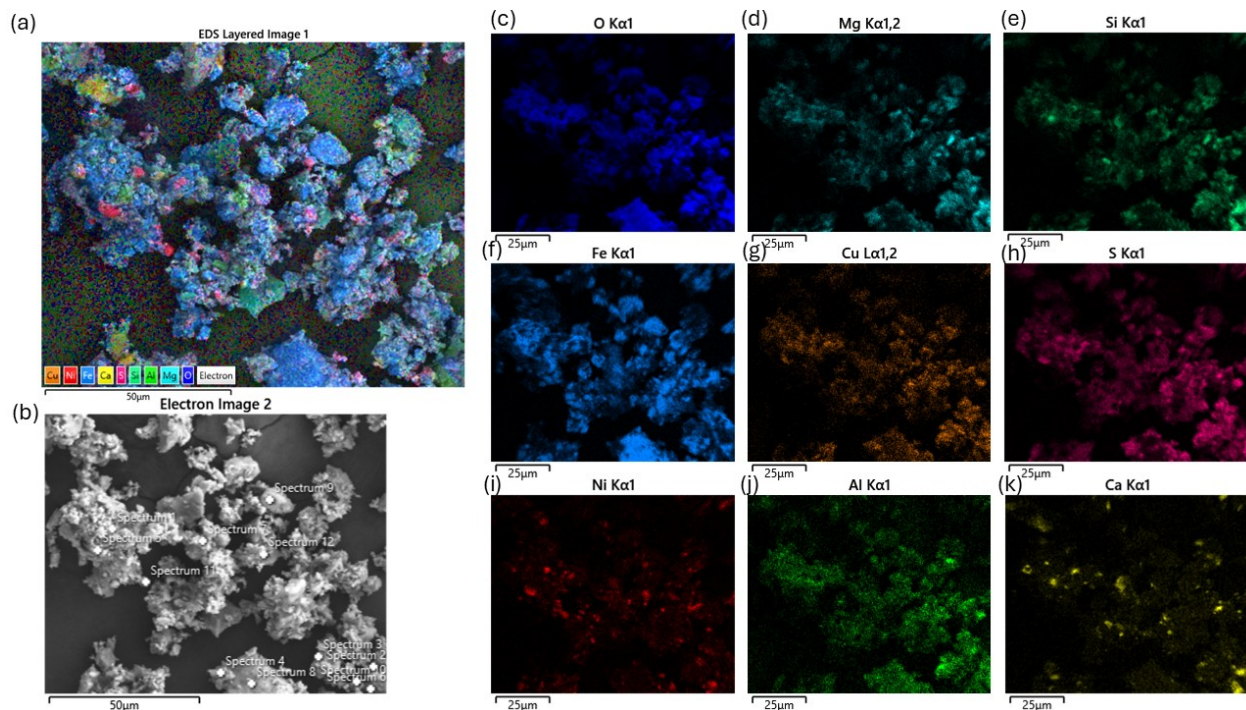


Figure 21: EDS mapping of roasted clinker with no salt additives

In Fig. 20 (b), the roasted clinkers with sodium sulfate exhibit distinct morphologies compared to the additive-free sample. The particles are more fragmented and porous, indicating enhanced phase transformation promoted by sodium sulfate. EDS analysis reveals that Ni occurs within Na–Fe–Ni–S complexes, signifying improved liberation of Ni compared with the additive-free condition. Cu is also detected in Fe–S complexes, suggesting partial co-extraction with Ni. Fe occurs both as oxides/sulfates and as components of Na–Fe–Ni–S complexes.

EDS mapping (Fig. 22) and Table S2 also demonstrate that some of the crystalline structures are unreacted Na_2SO_4 , implying incomplete reaction with mineral phases. The incomplete reaction could potentially be improved by ball milling the mixture to increase contact and reactivity compared to mortar mixing. While Mg-silicates remain similar to those in the no-additive case, the formation of Cu–Fe–Mg–silicate complexes suggest partial decomposition of the

serpentine-type gangue. Overall, the sodium sulfate additive enhances phase transformation and promotes Ni liberation from pentlandite.

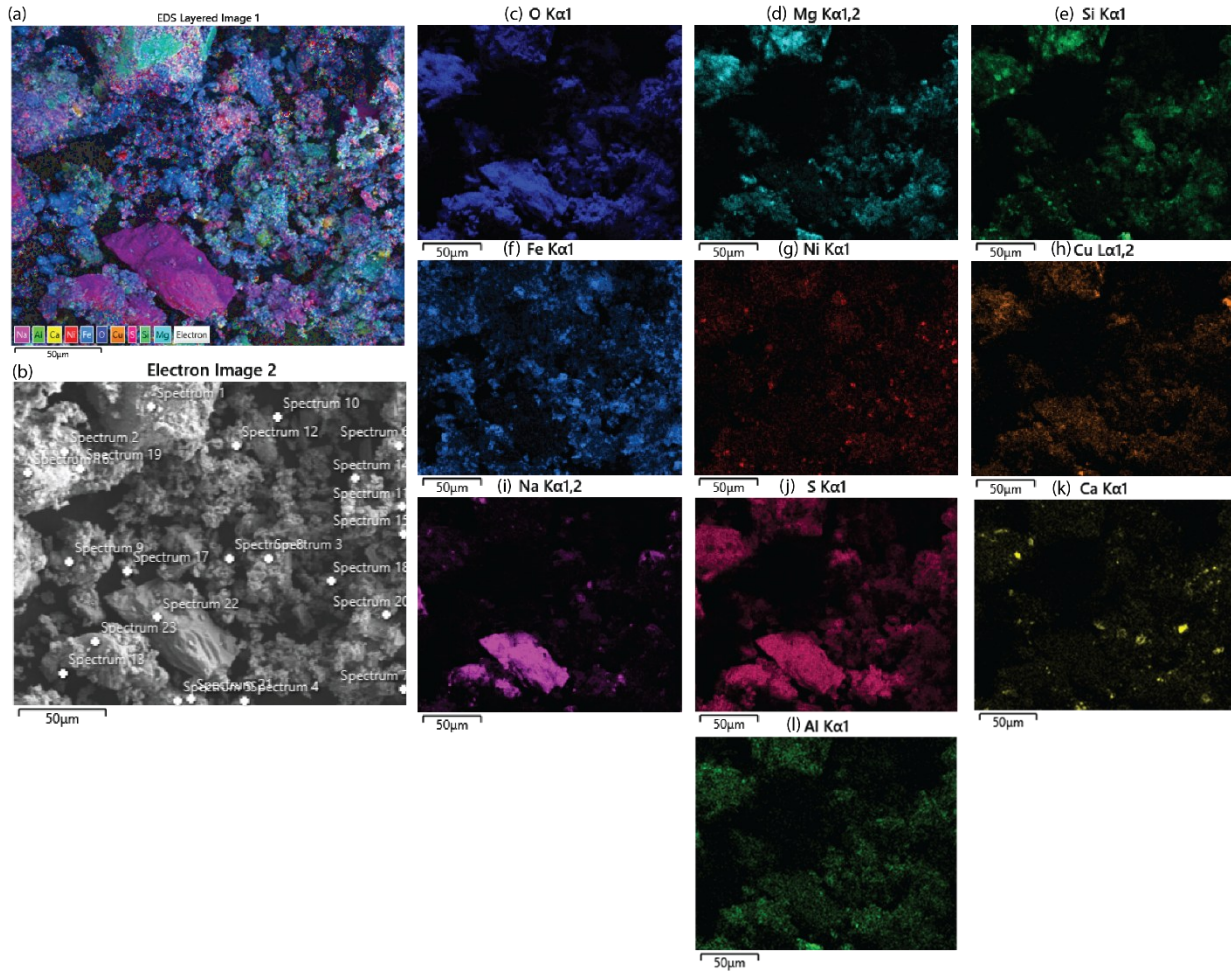


Figure 22: EDS mapping of roasted clinker using optimized sodium sulfate dosage

In Fig. 20(c), the roasted product with dual salt additives (sodium and potassium sulfate) exhibits pronounced morphological heterogeneity. The particles range from fine to coarse and display loosely aggregated, spongy, and porous structures. Such morphology is indicative of intensified sulfation and mineral phase transformations, which facilitate enhanced Ni liberation. In conjunction with the XRD analysis, EDS mapping (Fig. 23) and Table S3 also confirm the presence of double salts, showing increased incorporation of sulfur and alkali elements (Na, K) within the roasted clinkers, which signifies more extensive sulfation reactions as opposed to a single salt additive. However, Mg-silicate minerals, such as lizardite, remain partly intact, reflecting incomplete decomposition of serpentine-type gangue. Iron is frequently observed in association with Na, K, and S, suggesting the formation of jarosites, which is consistent with the XRD results. Despite these improvements, a fraction of Ni remains locked with Sulfur, as seen in XRD, thus suggesting the incomplete extraction. Similar to the single-salt system, unreacted sulfates are also present, with residual sodium sulfate being more prominent than potassium sulfate.

Overall, the dual-salt system demonstrates a synergistic effect: it promotes stronger sulfation reactions, improves mineral liberation, and consequently achieves significantly higher Ni

extraction efficiency compared to both the no-additive and single-additive systems.

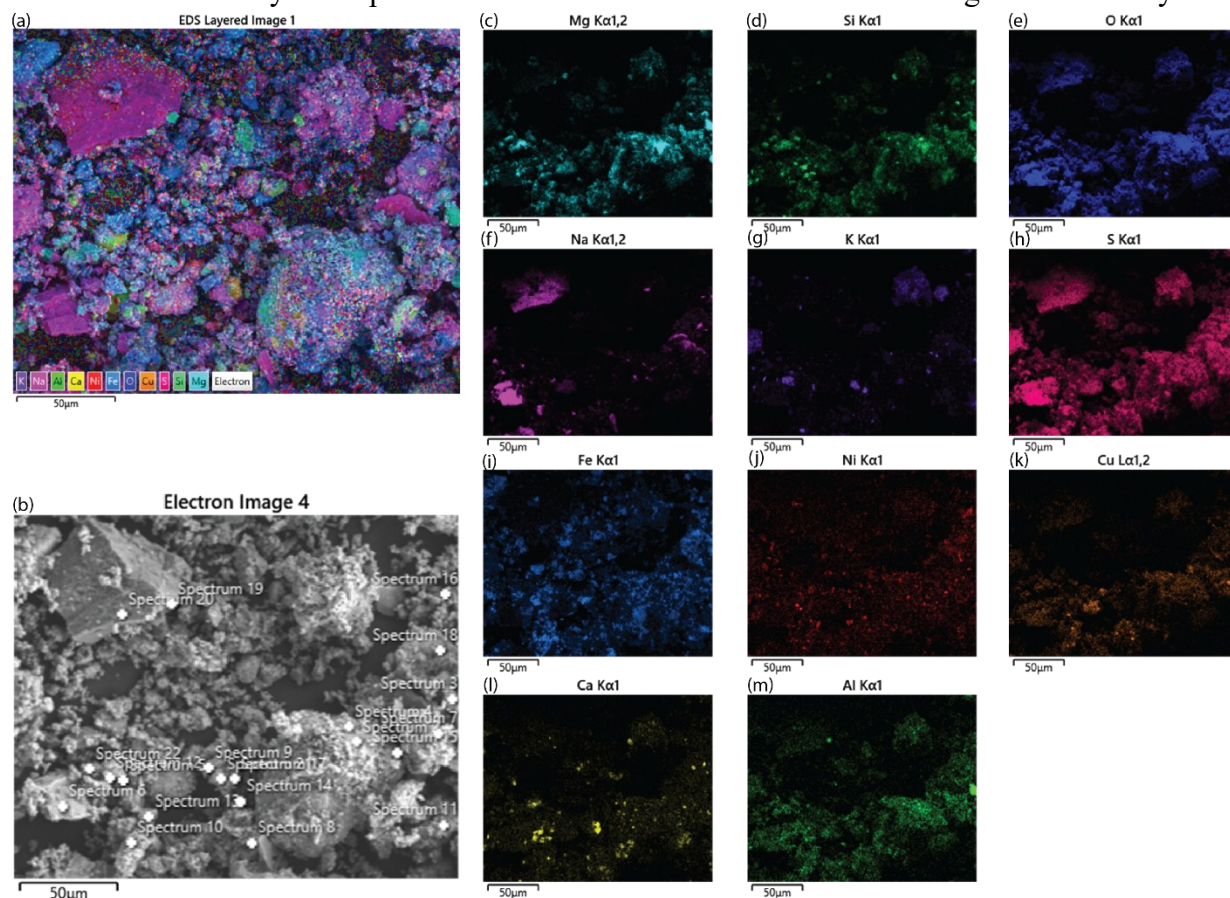


Figure 23: EDS mapping of roasted clinker with optimized dual salt dosage

Analysis of leached residue

The leached residue obtained after sulfation roasting under optimized process parameters was analyzed using SEM and XRD to evaluate the removal of impurities. The SEM micrographs (Fig. 24 (a) and (b)) reveal a heterogeneous mixture of coarse and fine particles exhibiting irregular shapes and porous morphology, indicative of structural changes induced during roasting and subsequent leaching. Complementary XRD analysis (Fig. 24 (c)) confirms that the predominant impurities removed as leach residue were iron-bearing phases. Specifically, ferric oxide (Fe_2O_3) and magnetite (Fe_3O_4) were identified as the dominant iron oxide species, with a certain amount of jarosites. The preferential oxidation of iron during the early stages of roasting facilitates the formation of stable oxide layers, which remain in the solid residue following leaching. This behavior highlights the high leaching selectivity of non-ferrous metals, allowing effective separation of nickel from iron impurities [2]. From a circular economy perspective, the recovery of Fe_2O_3 as a by-product is noteworthy. After suitable purification, this material can serve as a precursor for the synthesis of lithium iron phosphate (LFP) cathode active materials [11,12], thereby reducing waste generation and enhancing the sustainability of the process.

In addition to iron oxides, the residue also contained silicate gangue minerals, particularly serpentine-group phases such as lizardite ($\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$), which were also removed during the sulfation roasting process. A minor loss of nickel was also detected, primarily in the form of

residual NiS and NiO phases, suggesting incomplete leaching of nickel from the ore under the studied conditions.

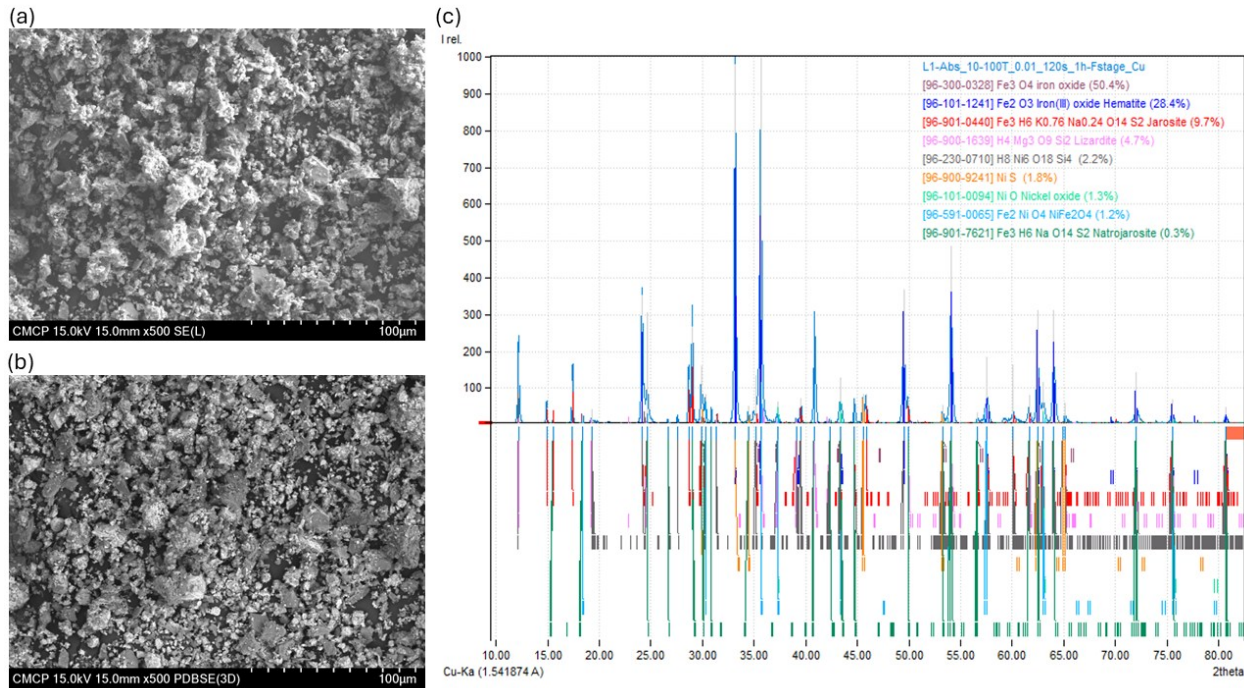


Figure 24: (a) and (b) SEM images of leached residue (c) XRD analysis of the leached residue

5.4. Concluding remarks and perspectives

This study successfully demonstrates the positive synergistic effect of a dual-salt system on nickel extraction, investigating potassium sulfate for the first time on nickel sulfide ore as part of an eco-friendly extraction approach. Unlike conventional methods that typically rely on concentrates, this study utilized direct ore containing high levels of impurities, showcasing the robustness of the method. Under the optimized roasting conditions: temperature (650 °C), time (2.5 h), sodium sulfate dosage (4 g), and potassium sulfate dosage (1.25 g), nickel extraction efficiency reached 79%, while 95% of iron was recovered in the leached residue. Notably, most of the recovered iron was present as Fe_2O_3 , which can serve as a precursor for the synthesis of LFP cathode active materials, highlighting the potential for valorizing the residue generated during Ni-rich extraction.

Despite the high extraction, some sodium and potassium sulfate remained unreacted in the roasted products. To enhance reaction kinetics and further improve nickel recovery, ball milling of the ore-salt mixture prior to roasting is recommended. Furthermore, achieving higher nickel extraction, potentially approaching 100%, could be realized through careful optimization of leaching parameters. In addition, incorporating environmentally friendly organic acids, such as citric acid, in controlled concentrations may provide additional benefits by forming chelates with nickel, thereby enhancing its extraction efficiency. Overall, this work not only establishes the efficacy of the dual-salt system for nickel sulfide ores but also provides actionable strategies for improving recovery and sustainability, paving the way for further studies on eco-friendly and efficient nickel extraction processes.

References

1. X. Xu et al., "Direct Extraction of Nickel and Copper from Low-Grade Nickel Sulfide Ore by Chlorination Roasting with Mixed $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ and NaCl ," JOM, vol. 74, no. 5, pp. 1989–1999, May 2022, doi: 10.1007/S11837-021-05122-X/TABLES/2.
2. Q. Sun et al., "Efficient Synchronous Extraction of Nickel, Copper, and Cobalt from Low-Nickel Matte by Sulfation Roasting–Water Leaching Process," Sci Rep, vol. 10, no. 1, pp. 1–14, Dec. 2020, doi: 10.1038/S41598-020-66894-X;SUBJMETA=440,638,639,905;KWRD=PHYSICAL+CHEMISTRY,PROCESS+CHEMISTRY.
3. Taylor & Francis, "Roasting," Taylor & Francis Knowledge Center, [Online]. Available: https://taylorandfrancis.com/knowledge/Engineering_and_technology/Materials_science/Roasting/. [Accessed: 18-Aug-2025].
4. K. Zhao, F. Gao, and Q. Yang, "Comprehensive Review on Metallurgical Upgradation Processes of Nickel Sulfide Ores," Journal of Sustainable Metallurgy, vol. 8, no. 1, pp. 37–50, Mar. 2022, doi: 10.1007/S40831-022-00501-3/FIGURES/7.
5. G. Li, H. Cheng, S. Chen, X. Lu, Q. Xu, and C. Lu, "Mechanism of Na_2SO_4 promoting nickel extraction from sulfide concentrates by sulfation roasting–water leaching," Metallurgical and Materials Transactions B, vol. 49, pp. 1136–1148, Apr. 2018, doi: 10.1007/s11663-018-1214-y
6. Mike, "Sodium sulfate price index," Business Analytiq, Oct. 22, 2024. [Online]. Available: <https://businessanalytiq.com/procurementanalytics/index/sodium-sulfate-price-index/>. [Accessed: 20-Jul-2025]
7. W. Mu, Z. Huang, H. Xin, S. Luo, Y. Zhai, and Q. Xu, "Extraction of copper and nickel from low-grade nickel sulfide ore by low-temperature roasting, selective decomposition and water-leaching process," JOM, vol. 71, pp. 4647–4658, Dec. 2019, doi: 10.1007/s11837-019-03740-0
8. W. Mu, F. Cui, Z. Huang, Y. Zhai, Q. Xu, and S. Luo, "Synchronous extraction of nickel and copper from a mixed oxide-sulfide nickel ore in a low-temperature roasting system," Journal of Cleaner Production, vol. 177, pp. 371–377, Mar. 2018, doi: 10.1016/j.jclepro.2017.12.260.
9. F. Cui, W. Mu, S. Wang, H. Xin, Q. Xu, Y. Zhai, and S. Luo, "Sodium sulfate activation mechanism on co-sulfating roasting to nickel-copper sulfide concentrate in metal extractions, microtopography and kinetics," Minerals Engineering, vol. 123, pp. 104–116, Oct. 2018, doi: 10.1016/j.mineng.2018.04.013
10. R. K. S. Hariyanto, L. Tomas da Rocha, S.-J. Kim, and S.-M. Jung, "Influence of alkali salts on extraction of nickel from serpentine-rich ore through sulfation roasting leaching process," Journal of Sustainable Metallurgy, vol. 9, pp. 1636–1646, Dec. 2023, doi: 10.1007/s40831-023-00753-7.
11. H. Rostami, J. Valio, P. Tynjälä, U. Lassi, and P. Suominen, "Life Cycle of LiFePO_4 Batteries: Production, Recycling, and Market Trends," ChemPhysChem, vol. 25, no. 24, art. no. e202400459, Dec. 2024, doi: 10.1002/cphc.202400459
12. A. Nekahi, A. K. M. R., X. Li, S. Deng, K. Zaghib, et al., "Sustainable LiFePO_4 and $\text{LiMn}_x\text{Fe}_{1-x}\text{PO}_4$ ($x = 0.1-1$) cathode materials for lithium-ion batteries: A systematic review from mine to chassis," Materials Science and Engineering: R: Reports, vol. 159, art. no. 100797, Jun. 2024, doi: 10.1016/j.mser.2024.100797.

Chapter 6 - Sustainable binders

6.1. Binders

Batteries consist of various components, such as electrodes, electrolytes, and current collectors. Most of the battery weight consists of these components, with binders and additives contributing a tiny percentage of the overall weight [1]. Since the invention of rechargeable battery technology, research and development efforts have primarily focused on electrode materials and electrolytes. Although binders are a critical component of batteries, they were typically ignored by battery engineers [2]. However, the rising demand for high-energy-density rechargeable batteries has brought attention to binders.

Binders are made of polymeric materials and account for approximately 5 wt% of the total battery weight [3]. Generally, they are electrochemically inactive. They serve multiple functions (Fig. 25): (1) providing stability to the battery; (2) ensuring homogeneous coating and adhesion of the active materials to the current collector; and (3) binding active materials and conductive additives together [4]. Binders are crucial for the electrode fabrication process. At the laboratory scale, binders are mixed with the electrode active material to form a slurry paste, which is subsequently coated onto the collector using the doctor blade technique and then dried [1]. Throughout the lithiation and delithiation processes, the volume changes in the electrode can damage its mechanical structure. Therefore, the binder used for electrode fabrication must possess sufficient mechanical properties, such as strength, elasticity, flexibility, and hardness, to maintain the integrity of the electrode. Furthermore, the lowest unoccupied molecular orbital (LUMO) of the binder used in the anode should be positioned higher than the chemical potential of the active material. Alternatively, the highest occupied molecular orbital (HOMO) of the binder should be lower than the chemical potential of the cathode [2].

As the battery market continues to expand, the demand for binders is expected to rise accordingly. The global binder market is projected to grow from USD 4.24 billion in 2025 to USD 8.81 billion by 2034, representing a CAGR of 8.46%. In 2024, Asia dominated the binder market with 40% of the market share, driven by countries including China, Japan, and South Korea [5]. Meanwhile, North America is experiencing significant growth in the binder market as well as in the battery industry overall. A significant driver of this growth is the increasing adoption of aqueous binders. In this thesis, emphasis is placed on sustainable aqueous binders, which have garnered significant attention in recent years due to their environmental compatibility and potential for large-scale implementation.

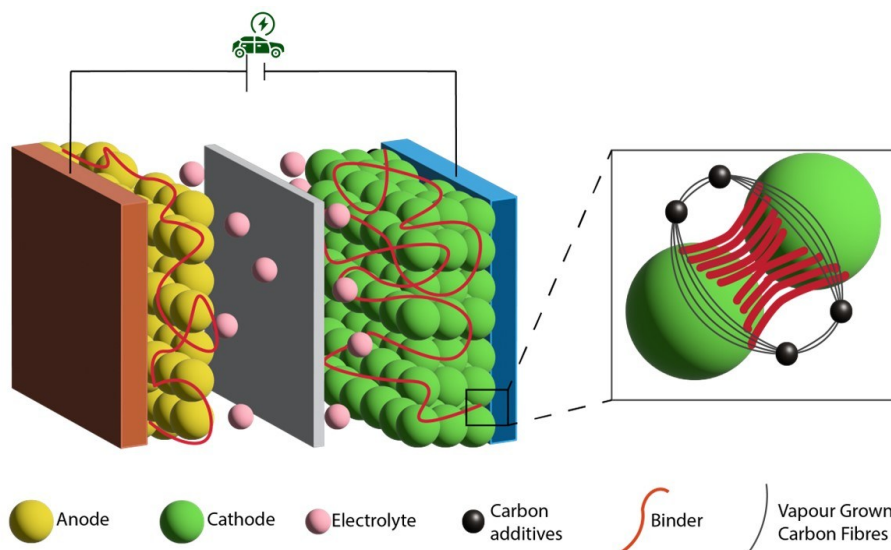


Figure 25: Illustrating the formation of cohesive networks by binders that anchor active material particles to the current collector, enhancing mechanical stability and maintaining electrical connectivity during cycling

6.2. Properties

6.2.1. Mechanical properties

The mechanical properties of binders, such as tensile strength, flexibility, elasticity, and adhesive strength, differ based on the polymer type, molecular structure, configuration, and functional groups [6]. These characteristics are crucial for the ability of an electrode to endure stresses caused by the expansion and contraction of the active materials during charge/discharge cycles. Strategies to achieve favorable mechanical properties in polymers often include improving intramolecular interactions or boosting crystallinity [7, 8].

6.2.1.1 Adhesion

Adhesive strength measures the bonding strength between the electrode film and the current collector [9]. This property is significantly affected by the molecular weight and functional groups of the polymer [2]. Chemical bonds, like hydrogen or ionic bonds, give binders, such as PAA and alginate, stronger adhesive forces compared to PVDF. Binders with high adhesive strength are crucial for maintaining solid connections among the electrode components, even during the volume expansion and contraction that occur during the charge/discharge cycle. A peeling test is performed to evaluate the adhesion strength of binders [7].

6.2.1.2. Tensile strength

The tensile strength represents the maximum tensile stress that a material can endure before breaking, indicating its resistance to mechanical failure [2, 9]. It measures the maximum force that can be withstood without failure and is directly linked to the structural stability provided by the polymer binder [10]. The chemical structure, molecular weight, and crystallinity of polymers influence this property. Higher molecular weight, increased crystallinity, greater crosslinking density, and stronger intermolecular interactions lead to higher tensile strength in the polymers [11, 12].

6.2.1.3. Elasticity and flexibility

The key characteristics of binders include elasticity and flexibility, which play a significant role in preserving a stable electrode structure in the face of naturally occurring volume changes in a battery [13, 14]. Flexibility refers to the ability of a material to bend without breaking, whereas elasticity refers to its ability to return to its former shape after being distorted. Binders exhibiting high levels of elasticity and flexibility, often polymers with low T_g , can better reduce electrode deformation caused by volume changes during charging and discharging cycles [7].

6.2.2. Electrical and ionic conductivity

Though conventional binders such as PVDF and CMC/SBR are electrically insulating, the incorporation of intrinsically conductive polymers (ICPs) such as PEDOT or polyaniline into electrode formulations can eliminate the need for separate conductive additives. Electrical conductivity in polymers is achieved through a π -conjugated network and free charge carriers [15]. In addition to electrical conductivity, the ionic conductivity of polymers is crucial for the electrochemical performance of batteries [16]. Factors, such as the degree of crystallinity, porosity, and viscosity of polymers, significantly affect their ionic conductivities. Other factors, such as the binder affinity with the electrolyte and the electrolyte wetting amount, are also critical in determining ionic conductivity. Structures with high porosity and amorphousness increase ionic conductivity [2]. Ionic conductivity can occur through specific mechanisms involving functional groups, such as Li-ion hopping [15].

6.2.3. Electrochemical and chemical stability

Binders must possess both chemical and electrochemical stability for prolonged periods and through multiple cycles without degrading the energy storage system [15]. To endure electrochemical reactions and prevent the corrosion of electrolytes during battery operation, binders need to be chemically stable (Fig. 26). The surrounding chemical environment, structure, and chemical composition affect stability [2]. Although byproducts from the chemically unstable binders can be used as components of the solid electrolyte interphase (SEI) layer on the electrode surface, excessive accumulation of these byproducts can reduce the coulombic efficiency (CE) and negatively impact cycling stability [7].

In terms of electrochemical stability, binders should have reversible redox activity or be redox-inactive, meaning that they should not oxidize or be reduced at high positive or low potentials. The electrochemical activity of binders can be beneficial for increasing the specific capacity of the electrodes [2]. X-ray photoelectron spectroscopy and Fourier-transform infrared spectroscopy can be used to evaluate these attributes [15].

6.2.4. Thermal stability

Thermal properties are critical for both the high-temperature conditions employed in electrode fabrication (such as coating and drying) and the operating conditions [15]. These properties encompass changes in the physical and chemical performance of polymers as heat is applied or removed. Thermal stability is influenced by various factors, such as the strength of binding forces among binders, their compositions, functional groups, primary and secondary structures, the kinetic motion of these structures, and molecular weight [17].

Although the typical operating temperature for polymer binders is below 55°C, they may be exposed to much higher temperatures, often exceeding 100°C, during manufacturing processes [18]. Furthermore, operational temperatures can unexpectedly increase, making high thermal

stability crucial over a wide temperature range [7]. In practice, electrodes must operate effectively in temperature ranges from -20 to 55°C [19]. Thus, the thermal properties of binders play a vital role in the fabrication and operation of energy-storage devices at elevated temperatures [2]. Thermogravimetric analysis is a technique used to measure thermal stability and determine the composition of materials [17].

6.2.5. Dispersion

The ability of polymers to uniformly spread the electrode components within the binder solution to produce a consistent composite is indicated by their dispersion properties. Uniform dispersion can be influenced by several factors, such as the polymer chain structure, charge density, chain flexibility, and mechanisms, such as electrostatic repulsion and depletion [2]. An uneven distribution of conductive additives, polymer binders, and active materials may have a detrimental effect on electron/ion conductivity, leading to elevated charge transfer resistances (R_{ct}) and localized current overloads [20]. Binders containing aromatic components can be beneficial for dispersing carbon-based conductive agents in electrodes due to their π – π stacking interactions, which facilitate effective dispersion [6].

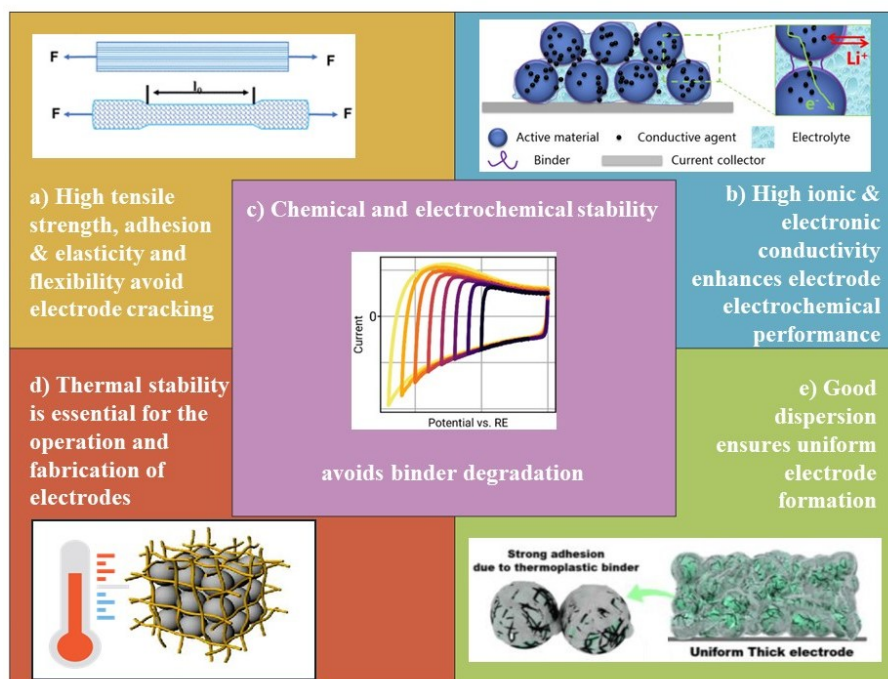


Figure 26: Illustration of the five critical properties associated with binders. (a) Mechanical properties, (b) electronic and ionic conductivity, (c) chemical and electrochemical stability, (d) thermal stability, and (e) dispersion properties. Reproduced with permission from [21-25]

6.3. Aqueous binders

Non-aqueous binders, such as PVDF, use organic dipolar aprotic solvents, N-Methyl-2-pyrrolidone (NMP), or polar aprotic solvents, dimethylformamide (DMF), which are highly hazardous to the environment. The use of NMP at the industrial scale is restricted by the Registration, Evaluation, and Authorization of Chemicals (REACH) regulation of the European Union [26]. To overcome the environmental challenges of non-aqueous binders, researchers have

focused on aqueous binders. Aqueous-based binders offer several advantages, such as cost-effectiveness, environmental friendliness, and less stringent processing requirements for air and humidity. Fig. 27. shows the advantages of aqueous binders over non-aqueous binders throughout the electrode preparation process. Additionally, they exhibit rapid solvent evaporation.

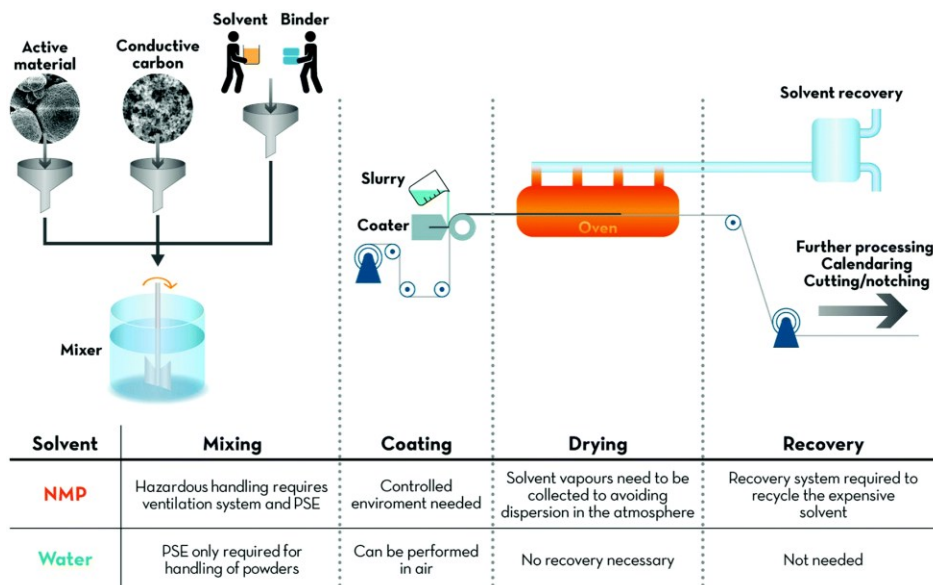


Figure 27: A simplified schematic representation of the production process for batteries. The primary benefits of using water instead of NMP are outlined for each stage, starting from the left and going rightward: the initial mixing of slurry, coating on the current collector, drying of the electrode, and solvent recovery. With permission from [27]

Compared with traditional PVDF-based binder electrodes, aqueous binder-based electrodes use a smaller quantity (approximately 5%) of the binders, thus enhancing the energy density of the cell. Another significant benefit of aqueous binders is their lower average swelling tendency in carbonate-based electrolytes compared to conventional non-aqueous binders. Furthermore, aqueous binders are more economical than their counterparts, making the fabrication process more cost-effective [1]. According to Wood et al., for a model LIB cell, processing electrodes with aqueous binders is 26 times less expensive than using the conventional NMP-based method, primarily due to the substantial cost savings from omitting the energy-intensive solvent recovery process. This economic advantage, along with environmental benefits, is driving the shift towards aqueous binder systems in the battery industry [28]. Zeon Co., Ltd. is a pioneering Japanese company in the development of water-based binders [4]. Over the years, several aqueous binders have been explored, including polytetrafluoroethylene (PTFE), styrene-butadiene rubber (SBR), chitosan, polyacrylic acid (PAA), and sodium alginate (SA), as shown in Fig. 28.

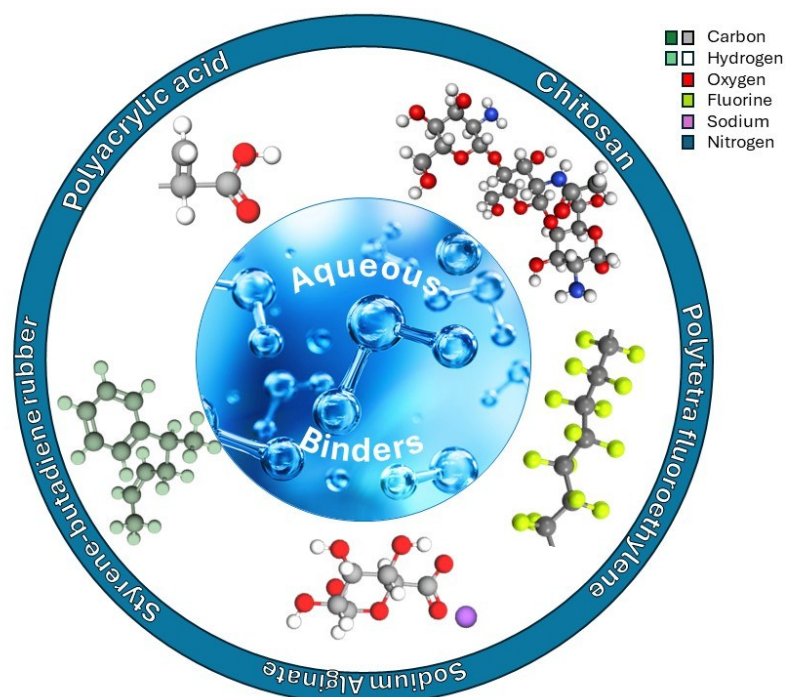


Figure 28: Illustration of the various aqueous binders available in the market for rechargeable batteries

6.3.1. Polytetrafluoroethylene (PTFE)

PTFE is recognized for its environmental friendliness, as it uses water as a solvent and poses no health risks. It has high chemical resistance, high hydrophobicity, a wide temperature range, insulation properties, UV resistance, weather resistance, low friction coefficient, fracture and flexural resistance, fire resistance, and minimal water absorption [29]. These properties result from its high crystallinity, molecular weight, and unbranched structure, characterized by significant C-F bonding energy (485 kJ mol^{-1}) [30, 31]. Although PTFE is associated with a capacity loss in carbon electrodes, these characteristics make it a better binder compared to PVDF. Owing to its superior crystallinity, high post-sintering percentage ($>97\%$), and fiber-forming capacity, it can be considered a suitable binder, especially for LIBs [32].

Compared with other polymers, PTFE is less resilient and more flexible. It also has moderate tensile strength and high elongation at break, which makes it appropriate for solvent-free processes and polymer fibrillation techniques [30, 31]. The fibrillation characteristics of PTFE aqueous dispersions offer an excellent binding capacity with electrode materials, although excessive fibrillation may prevent homogeneous dispersion, leading to poor interconnection with electrodes. The incompatibility of active materials with water and stabilizers presents further challenges, as the PTFE binder relies on an aqueous solvent and stabilizer for the electrode construction process [4, 33]. Gao et al. used PTFE as an aqueous binder instead of the organic binder PVDF to fabricate an LFP/C cathode. The cell incorporating the PTFE binder demonstrated an increased discharge capacity of 161.1 mAh g^{-1} and a lower shear rate compared with PVDF. Notably, the interfacial resistance (R_{ct}) of the PTFE cell was lower compared to that of PVDF, with values of 21.4 and $28.9 \text{ } \Omega$, respectively [34].

6.3.2. Styrene-Butadiene Rubber (SBR)

SBR is produced through the synthesis of 1,3-butadiene and styrene monomers, and its characteristics depend on its molecular structure, including the ratio of styrene to butadiene, degree of cross-linking (DoCL), type of emulsifier, and polymerization temperature [1, 35]. Compared with PVDF, SBR is a better elastomer because it is more flexible, has better binding properties, and can withstand higher temperatures [36]. Zeon Co., Ltd. is a prominent manufacturer of SBR binders for both anodes and cathodes [1].

SBR is generally used as a binder in combination with carboxymethyl cellulose (CMC). Within the SBR/CMC system, SBR acts as a binder, while CMC regulates the dispersibility and viscosity of the slurry [37]. In graphite anodes, SBR provides flexibility to the SBR/CMC system, which tends to become brittle after vacuum drying. The SBR/CMC binder improves the adhesion between the electrode film and current collector, forming an efficient three-dimensional network for electron transport. The SBR/CMC binder also aids in creating a uniform SEI film, thereby preventing the formation of Li dendrites. Zhang et al. used a mixture of SBR/CMC in weight ratios of 1:1, 1:3, and 3:1 as a binder for a ZnFe_2O_4 (ZFO) electrode in LIBs. The 1:1 configuration achieved a discharge capacity of 873.8 mAh g^{-1} after 100 cycles at a 0.1 C rate, exhibiting a minimal capacity fading rate of 0.06% per cycle. Thus, indicating the potential of the SBR/CMC binder [38]. However, it has been observed that the SBR/CMC system fails with the Si anode due to its low binding ability concerning the volume expansion of the Si anode [39].

The DoCL and the ratio of styrene to butadiene units are two important factors that affect the properties of SBR, including its tensile strength and T_g [37]. To understand the effect of the DoCL on the properties of the SBR/CMC binder, Isozumi et al. conducted a study on LCO composite electrodes by modifying the DoCL within SBRs. They found that SBRs with a lower DoCL exhibited better electrochemical properties [40]. Conversely, Jolley et al. examined the effects of different DoCLs in SBRs on graphite anodes using a CMC/SBR binder system. Their results showed that SBRs with a higher DoCL provide better and more consistent capacity retention than those with a lower DoCL. A higher DoCL content in SBRs also resulted in electrode coatings with stronger cohesiveness, which reduced active material loss and capacity fading over time by reducing delamination between the active material layers [41].

SBR/CMC is widely used as an aqueous binder due to its strong mechanical properties, making it a preferred choice. When combined with CMC, these polymers work together effectively to meet current binder requirements. Additionally, it has the potential to replace environmentally harmful PVDF in the market.

6.3.3. Polyacrylic acid (PAA)

PAA serves as a functional polymeric binder featuring carboxylic acid functional groups ($-\text{COOH}$) that establish weak hydrogen bonds with the surface $-\text{OH}$ groups of the active materials, thereby forming a conductive ionic film [4, 42, 43]. As a binder, PAA imparts advantages, such as robust binding ability, adhesive strength, swelling properties, high stress at break (90 MPa), and uniform particle distribution within the electrode [44]. The electrical properties of PAA improve with the increase in molecular weight and higher concentrations of Li^+ , enhancing its overall performance [45, 46]. For PAA, a high pH value improves particle dispersion but severely compromises the mechanical properties of the binder [46]. PAA is soluble in both water and NMP, facilitating strong hydrogen bond formation with active materials and current collectors because of the presence of carboxylic acid functional groups [44]. PAA also enables regulation of the distance

between functional groups by copolymerization with other monomers, thereby facilitating the adjustment of the mechanical characteristics of the binder and its ability to swell and dissolve in an electrolyte solvent [47]. In addition, PAA binders help graphite surfaces establish a consistent and stable SEI layer that effectively blocks direct contact between graphite and the electrolyte, thereby reducing the rate at which the electrolyte decomposes [48]. Moreover, for silicon-based anodes, PAA demonstrates exceptional cycling performance compared with CMC or PVDF [47].

Extensive studies have explored the use of PAA as an aqueous binder in LFP cathodes. The obtained results indicated reduced polarization, lower resistance, and higher reversibility with sustained capacity retention (99.3%) for up to 200 cycles compared to LFP/PVDF cathodes [49]. However, PAA is less effective in lithium nickel manganese oxide (LNMO) electrodes with high mass loadings owing to specific challenges, such as high internal resistance in the initial cycles, which is attributed to several factors, such as contact resistance, inhomogeneous binder distribution, and poor electrolyte wetting. By modifying the polymer chemistry of PAA, problems such as agglomeration and electrode pore clogging can be reduced. This modification can be achieved by copolymerizing PAA with various functional groups, which can enhance specific properties, such as adhesion strength, surface chemistry, and electrolyte swelling, thus improving the potential of the PAA binder system for LNMO [50].

6.3.4. Chitosan

Chitosan, primarily composed of β -(1,4)-linked 2-deoxy-2-amino-D-glucopyranose units, is derived from chitin, a natural biopolymer derived from the partial deacetylation of poly(β -(1 $\rightarrow$ 4)-N-acetyl-D-glucosamine) [51, 52]. Chitin has a structure similar to that of cellulose, but with the hydroxyl groups replaced by acetamide groups ($-\text{NHCOCH}_3$) [53]. Unlike chitin, chitosan is soluble in dilute aqueous acetic acid solutions, in which the glucosamine unit ($\text{R}-\text{NH}_2$) protonates into its positively charged form ($\text{R}-\text{NH}^{3+}$) [54, 55]. Its abundance and the presence of amino and hydroxyl functionalities have rendered chitosan conducive for copolymerization or hybridization with other functional polymers, making it an effective water-soluble binder for various electrode materials [1]. Chitosan was initially introduced as a binder by Chai et al. for a graphite anode, achieving a CE of up to 95.4%. Chitosan has been applied in various aspects of LIBs, serving as a template during active material synthesis, a precursor for carbon coating, a binder for numerous anode materials, and gel polymer electrolyte [51].

One study explored the biopolymer chitosan as a promising binder for producing LFP cathodes in LIBs. Utilizing chitosan as a binder resulted in an electrode with a high discharge capacity of 159.4 mAh g^{-1} , along with an outstanding capacity retention ratio of 98.38% [56]. Several properties dictate the use of chitosan as a binder. The degree of acetylation (DA), which is the percentage of monomeric acetylated units, and the degree of polymerization (DP), which is the number of monomer units in a biopolymer, are crucial parameters that dictate the chemical and physical properties of chitin and chitosan [57]. Hamzelui et al. evaluated chitosan samples with various DAs and DPs as binders for Si/Gr-based anodes. Electrochemical measurements conducted in Si/Gr||Li metal and NMC622||Si/Gr full cells led to the selection of a chitosan binder with a high DP of 1618 and a DA of 50% (DA50) as the optimized version for evaluating Si/Gr anodes in battery cells [52].

6.3.5. Sodium Alginate (SA)

Alginate, a high-modulus polysaccharide, is widely distributed in the cell walls of brown algae and other microorganisms [1]. It comprises copolymers of β -D-mannuronate units (M block)

and α -L-glucuronate (G block) linked by 1,4 linkages. Alginate benefits from the presence of metal ions, which enhances its mechanical properties [58, 59]. Owing to the abundant carboxyl groups in its polymer chains, SA forms strong interactions, such as hydrogen or covalent bonds, with active material surfaces [1]. It dissociates readily in water to generate carboxylic anionic free radicals [60]. SA also exhibits excellent mechanical properties in electrolyte solvents, boasting a stiffness that is approximately 50 times higher than that of PVDF [61].

When used as a binder mixed with a Si anode, alginate demonstrated remarkable electrochemical capacity improvement and cycling stability compared to CMC or PVDF binders [62]. The reversible and self-healing bonds formed in the presence of calcium (Ca^{2+}) or magnesium (Mg^{2+}) ions contribute to extreme toughness and resilience. Alginate, particularly when crosslinked with Ca^{2+} , significantly enhances the cycle stability of Si-based anodes on Cu-foil current collectors [63, 64, 65]. The G blocks of alginate specifically bind with Ca^{2+} to create a series of reversible and robust electrostatic crosslinks known as the “egg-box structure”. The calcium-mediated “egg-box” electrostatic crosslinking of alginate enhances toughness, resilience, and electrolyte desolvation, thus improving its performance as an Si-binder for LIBs, as observed by Yoon et al. [66].

6.4. Concluding remarks and perspectives

The binder plays a crucial role in LIBs by ensuring strong adhesion between the active material and the current collector. Without adequate binding, delamination can occur during electrode fabrication or cycling, leading to compromised electrochemical performance. As the demand for sustainable LIBs continues to grow, the development and adoption of environmentally benign components, such as aqueous binders, are essential. Promoting aqueous binder systems thus represents a significant step toward greener battery technologies.

Several aqueous binders have already been investigated and adopted commercially for both positive and negative electrodes. Among them, the SBR/CMC system is widely used, particularly in graphite anodes, and has demonstrated superior electrochemical performance and processing advantages over conventional PVDF, thereby highlighting the clear potential of aqueous binders to replace toxic, solvent-based binders.

Although most aqueous binders are electrochemically inactive, the integration of ICPs offers a promising strategy to enhance specific capacity by eliminating the need for additional conductive additives. This multifunctionality can further improve electrode performance while simplifying electrode design.

To comprehensively validate the environmental credentials of aqueous binders, LCA studies are essential. Research has established the cost-effectiveness of aqueous binders, mainly due to the elimination of energy-intensive solvent recovery steps. By employing LCA for binders, the quantification of broader environmental benefits can be achieved. Demonstrating both economic and environmental advantages through such assessments is critical to solidifying aqueous binders as a commercially viable and sustainable alternative to PVDF in large-scale LIB production.

References

1. N. Lingappan, L. Kong, and M. Pecht, "The significance of aqueous binders in lithium-ion batteries," *Renewable and Sustainable Energy Reviews*, vol. 147, p. 111227, Apr. 2021, doi: 10.1016/j.rser.2021.111227.
2. H. Chen, M. Ling, L. Hencz, H. Y. Ling, G. Li, Z. Lin, G. Liu, and S. Zhang, "Exploring Chemical, Mechanical, and Electrical Functionalities of Binders for Advanced Energy-Storage Devices," Sep. 26, 2018, American Chemical Society. doi: 10.1021/acs.chemrev.8b00241
3. F. Jeschull, J. Maibach, K. Edström, and D. Brandell, "On the Electrochemical Properties and Interphase Composition of Graphite: PVdF-HFP Electrodes in Dependence of Binder Content," *J Electrochem Soc*, vol. 164, no. 7, pp. A1765–A1772, Jun. 2017, doi: 10.1149/2.0121709jes.
4. Y. Ma, J. Ma, and G. Cui, "Small things make big deal: Powerful binders of lithium batteries and post-lithium batteries," *Energy Storage Mater*, vol. 20, pp. 146–175, Jul. 2019, doi: 10.1016/J.ENSMS.2018.11.013.
5. Precedence Research, "Battery binders market size, trends, and forecast 2024 to 2032," Precedence Research, 2024. [Online]. Available: <https://www.precedenceresearch.com/battery-binders-market>
6. Y. Yang, S. Wu, Y. Zhang, C. Liu, X. Wei, D. Luo, and Z. Lin, "Towards efficient binders for silicon based lithium-ion battery anodes," *Chemical Engineering Journal*, vol. 406, p. 126807, Feb. 2021, doi: 10.1016/j.cej.2020.126807.
7. H.-J. Jin, J. Lee, J. Yoon, H. Kim, and J. Kim, "Polymeric Binder Design for Sustainable Lithium-ion Battery Chemistry," Dec. 2023, doi: 10.20944/PREPRINTS202312.0863.V1.
8. U. S. Vogl, P. K. Das, A. Z. Weber, M. Winter, R. Kostecki, and S. F. Lux, "Mechanism of Interactions between CMC Binder and Si Single Crystal Facets," *Langmuir*, vol. 30, no. 34, pp. 10299–10307, Sep. 2014, doi: 10.1021/la501791q.
9. J. E. Mark, Ed., *Physical Properties of Polymers Handbook*. New York, NY: Springer New York, 2007. doi: 10.1007/978-0-387-69002-5.
10. Q. Zhang, J. Chen, B. Jin, and R. Peng, "A new linear heptafluoro glycidyl ether binder: synthesis, characterization, and mechanical properties," *Macromol Res*, vol. 31, no. 7, pp. 699–709, Jul. 2023, doi: 10.1007/s13233-023-00162-5.
11. J. Liu, D. G. D. Galpaya, L. Yan, M. Sun, Z. Lin, C. Yan, C. Liang, and S. Zhang, "Exploiting a robust biopolymer network binder for an ultrahigh-areal-capacity Li-S battery," *Energy Environ Sci*, vol. 10, no. 3, pp. 750–755, 2017, doi: 10.1039/C6EE03033E
12. D. Shin, H. Park, and U. Paik, "Cross-linked poly(acrylic acid)-carboxymethyl cellulose and styrene-butadiene rubber as an efficient binder system and its physicochemical effects on a high energy density graphite anode for Li-ion batteries," *Electrochem commun*, vol. 77, pp. 103–106, Apr. 2017, doi: 10.1016/j.elecom.2017.02.018
13. J. Ahn, H. G. Im, Y. Lee, D. Lee, H. Jang, Y. Oh, K. Chung, T. Park, M. K. Um, J. W. Yi, and J. Kim, "A novel organosilicon-type binder for LiCoO₂ cathode in Li-ion batteries," *Energy Storage Mater*, vol. 49, pp. 58–66, Aug. 2022, doi: 10.1016/j.ensm.2022.04.005
14. Y. Shi, F. Gao, Y. Xie, X. Xu, F. Li, X. Han, X. Yao, D. Wang, Y. Hou, X. Gao, and Q. He, "In situ interlocked gradient adaptive network binder with robust adhesion and cycle performance for silicon anodes," *J Power Sources*, vol. 580, p. 233267, Oct. 2023, doi: 10.1016/j.jpowsour.2023.233267.

15. A. Cholewinski, P. Si, M. Uceda, M. Pope, and B. Zhao, "Polymer Binders: Characterization and Development toward Aqueous Electrode Fabrication for Sustainability," *Polymers (Basel)*, vol. 13, no. 4, p. 631, Feb. 2021, doi: 10.3390/polym13040631
16. C. Sun, J. Liu, Y. Gong, D. P. Wilkinson, and J. Zhang, "Recent advances in all-solid-state rechargeable lithium batteries," *Nano Energy*, vol. 33, pp. 363–386, Mar. 2017, doi: 10.1016/j.nanoen.2017.01.028.
17. A. V. Le, M. Wang, D. J. Noelle, Y. Shi, Y. S. Meng, D. Wu, J. Fan, and Y. Qiao, "Using high-HFP-content cathode binder for mitigation of heat generation of lithium-ion battery," *Int J Energy Res*, vol. 41, no. 14, pp. 2430–2438, Nov. 2017, doi: 10.1002/er.3824.
18. Y. Komoda, K. Ishibashi, K. Kuratani, K. Suzuki, N. Ohmura, and H. Kobayashi, "Effects of drying rate and slurry microstructure on the formation process of LiB cathode and electrochemical properties," *J Power Sources*, vol. 568, p. 232983, Jun. 2023, doi: 10.1016/j.jpowsour.2023.232983.
19. L. Lu, X. Han, J. Li, J. Hua, and M. Ouyang, "A review on the key issues for lithium-ion battery management in electric vehicles," *J Power Sources*, vol. 226, pp. 272–288, Mar. 2013, doi: 10.1016/j.jpowsour.2012.10.060.
20. W. Zeng, L. Wang, X. Peng, T. Liu, Y. Jiang, F. Qin, L. Hu, P. K. Chu, K. Huo, and Y. Zhou, "Enhanced Ion Conductivity in Conducting Polymer Binder for High-Performance Silicon Anodes in Advanced Lithium-Ion Batteries," *Adv Energy Mater*, vol. 8, no. 11, Apr. 2018, doi: 10.1002/aenm.201702314.
21. L. Zhang, X. Wu, W. Qian, K. Pan, X. Zhang, L. Li, M. Jia, and S. Zhang, "Exploring More Functions in Binders for Lithium Batteries," *Electrochemical Energy Reviews* 2023 6:1, vol. 6, no. 1, pp. 1–44, Dec. 2023, doi: 10.1007/S41918-023-00198-2.
22. Lim, S.; Chu, H.; Lee, K.; Yim, T.; Kim, Y.J.; Mun, J.; Kim, T.H, "Physically Cross-linked Polymer Binder Induced by Reversible Acid–Base Interaction for High-Performance Silicon Composite Anodes," *ACS Appl Mater Interfaces*, vol. 7, no. 42, pp. 23545–23553, Oct. 2015, doi: 10.1021/acsami.5b06682.
23. H.-M. Kim, B.-I. Yoo, J.-W. Yi, M.-J. Choi, and J.-K. Yoo, "Solvent-Free Fabrication of Thick Electrodes in Thermoplastic Binders for High Energy Density Lithium-Ion Batteries," *Nanomaterials*, vol. 12, no. 19, p. 3320, Sep. 2022, doi: 10.3390/nano12193320.
24. P. Ruschhaupt, S. Pohlmann, A. Varzi, and S. Passerini, "Determining Realistic Electrochemical Stability Windows of Electrolytes for Electrical Double-Layer Capacitors," *Batter Supercaps*, vol. 3, no. 8, pp. 698–707, Aug. 2020, doi: 10.1002/batt.202000009
25. Liang, X.; Ahmad, N.; Zhang, B.; Zeng, C.; Cao, X.; Dong, Q.; Yang, W, "Research progress of robust binders with superior mechanical properties for high-performance silicon-based lithium-ion batteries," *Mater Chem Front*, vol. 8, no. 6, pp. 1480–1512, 2024, doi: 10.1039/D3QM00839H.
26. Y. Bai, W. B. Hawley, C. J. Jafta, N. Muralidharan, B. J. Polzin, and I. Belharouak, "Sustainable recycling of cathode scraps via Cyrene-based separation," *Sustainable Materials and Technologies*, vol. 25, p. e00202, Sep. 2020, doi: 10.1016/j.susmat.2020.e00202.
27. D. Bresser, D. Buchholz, A. Moretti, A. Varzi, and S. Passerini, "Alternative binders for sustainable electrochemical energy storage – the transition to aqueous electrode processing and bio-derived polymers," *Energy Environ Sci*, vol. 11, no. 11, pp. 3096–3127, 2018, doi: 10.1039/C8EE00640G

28. D. L. Wood, J. Li, and C. Daniel, "Prospects for reducing the processing cost of lithium ion batteries," *J Power Sources*, vol. 275, pp. 234–242, Feb. 2015, doi: 10.1016/j.jpowsour.2014.11.019.
29. S. Priyono, B. M. Lubis, S. Humaidi, and B. Prihandoko, "Heating Effect on Manufacturing Li 4 Ti 5 O 12 Electrode Sheet with PTFE Binder on Battery Cell Performance," *IOP Conf Ser Mater Sci Eng*, vol. 367, p. 012007, May 2018, doi: 10.1088/1757-899X/367/1/012007
30. P. J. Rae and E. N. Brown, "The properties of poly(tetrafluoroethylene) (PTFE) in tension," *Polymer (Guildf)*, vol. 46, no. 19, pp. 8128–8140, Sep. 2005, doi: 10.1016/j.polymer.2005.06.120.
31. L. A. Pruitt, "Deformation, yielding, fracture and fatigue behavior of conventional and highly cross-linked ultra high molecular weight polyethylene," *Biomaterials*, vol. 26, no. 8, pp. 905–915, Mar. 2005, doi: 10.1016/j.biomaterials.2004.03.022.
32. E. N. Brown, P. J. Rae, E. Bruce Orler, G. T. Gray, and D. M. Dattelbaum, "The effect of crystallinity on the fracture of polytetrafluoroethylene (PTFE)," *Materials Science and Engineering: C*, vol. 26, no. 8, pp. 1338–1343, Sep. 2006, doi: 10.1016/j.msec.2005.08.009.
33. X. Wang, S. Chen, K. Zhang, L. Huang, H. Shen, Z. Chen, C. Rong, G. Wang, and Z. Jiang, "A Polytetrafluoroethylene-Based Solvent-Free Procedure for the Manufacturing of Lithium-Ion Batteries," *Materials*, vol. 16, no. 22, p. 7232, Nov. 2023, doi: 10.3390/ma16227232.
34. S. Gao, Y. Su, L. Bao, N. Li, L. Chen, Y. Zheng, J. Tian, J. Li, S. Chen, and F. Wu, "High-performance LiFePO₄/C electrode with polytetrafluoroethylene as an aqueous-based binder," *J Power Sources*, vol. 298, pp. 292–298, Dec. 2015, doi: 10.1016/j.jpowsour.2015.08.074.
35. K. S. Sisanth, M. G. Thomas, J. Abraham, and S. Thomas, "General introduction to rubber compounding," in *Progress in Rubber Nanocomposites*, Elsevier, 2017, pp. 1–39. doi: 10.1016/B978-0-08-100409-8.00001-2
36. M. Yoshio, S. Kugino, and N. Dimov, "Electrochemical behaviors of silicon based anode material," *J Power Sources*, vol. 153, no. 2, pp. 375–379, Feb. 2006, doi: 10.1016/j.jpowsour.2005.05.052.
37. Park, K.; Yoo, H.E.; Jung, Y.; Ryu, M.; Myeong, S.; Lee, D.; Kim, S.C.; Kim, C.; Kim, J.; Kwon, J.; Lee, K, "Styrene-butadiene rubber patterned current collector for improved Li-ion kinetics of the anode for high energy density lithium-ion batteries," *J Power Sources*, vol. 577, p. 233238, Sep. 2023, doi: 10.1016/j.jpowsour.2023.233238.
38. R. Zhang, X. Yang, D. Zhang, H. Qiu, Q. Fu, H. Na, Z. Guo, F. Du, G. Chen, and Y. Wei, "Water soluble styrene butadiene rubber and sodium carboxyl methyl cellulose binder for ZnFe₂O₄ anode electrodes in lithium ion batteries," *J Power Sources*, vol. 285, pp. 227–234, Jul. 2015, doi: 10.1016/j.jpowsour.2015.03.100.
39. Y.-M. Zhao, F.-S. Yue, S.-C. Li, Y. Zhang, Z.-R. Tian, Q. Xu, S. Xin, and Y.-G. Guo, "Advances of polymer binders for <scp>silicon-based</scp> anodes in high energy density <scp>lithium-ion</scp> batteries," *InfoMat*, vol. 3, no. 5, pp. 460–501, May 2021, doi: 10.1002/inf2.12185.
40. H. Isozumi, T. Horiba, K. Kubota, K. Hida, T. Matsuyama, S. Yasuno, and S. Komaba, "Application of modified styrene-butadiene-rubber-based latex binder to high-voltage operating LiCoO₂ composite electrodes for lithium-ion batteries," *J Power Sources*, vol. 468, p. 228332, Aug. 2020, doi: 10.1016/j.jpowsour.2020.228332
41. M. J. Jolley, T. S. Pathan, C. Jenkins, and M. J. Loveridge, "Investigating the effect of the degree of cross-linking in styrene butadiene rubber on the performance of graphite anodes

- for the use in lithium-ion batteries,” *J Appl Polym Sci*, vol. 141, no. 12, Mar. 2024, doi: 10.1002/app.55135.
42. Z. Ma, Y. Lyu, H. Yang, Q. Li, B. Guo, and A. Nie, “Systematic investigation of the Binder’s role in the electrochemical performance of tin sulfide electrodes in SIBs,” *J Power Sources*, vol. 401, pp. 195–203, Oct. 2018, doi: 10.1016/j.jpowsour.2018.08.081.
 43. P. Parikh, M. Sina, A. Banerjee, X. Wang, M. S. D’Souza, J. M. Doux, E. A. Wu, O. Y. Trieu, Y. Gong, Q. Zhou, and K. Snyder, “Role of Polyacrylic Acid (PAA) Binder on the Solid Electrolyte Interphase in Silicon Anodes,” *Chemistry of Materials*, vol. 31, no. 7, pp. 2535–2544, Apr. 2019, doi: 10.1021/acs.chemmater.8b05020.
 44. Z. Zhang, T. Zeng, Y. Lai, M. Jia, and J. Li, “A comparative study of different binders and their effects on electrochemical properties of LiMn_2O_4 cathode in lithium ion batteries,” *J Power Sources*, vol. 247, pp. 1–8, Feb. 2014, doi: 10.1016/j.jpowsour.2013.08.051.
 45. Hu, B.; Shkrob, I.A.; Zhang, S.; Zhang, L.; Zhang, J.; Li, Y.; Liao, C.; Zhang, Z.; Lu, W.; Zhang, L, “The existence of optimal molecular weight for poly(acrylic acid) binders in silicon/graphite composite anode for lithium-ion batteries,” *J Power Sources*, vol. 378, pp. 671–676, Feb. 2018, doi: 10.1016/j.jpowsour.2017.12.068.
 46. F. Sun and D. R. Wheeler, “The Effects of Lithium Ions and pH on the Function of Polyacrylic Acid Binder for Silicon Anodes,” *J Electrochem Soc*, vol. 170, no. 8, p. 080502, Aug. 2023, doi: 10.1149/1945-7111/aceab1.
 47. Magasinski, A.; Zdyrko, B.; Kovalenko, I.; Hertzberg, B.; Burtovyy, R.; Huebner, C.F.; Fuller, T.F.; Luzinov, I.; Yushin, G, “Toward Efficient Binders for Li-Ion Battery Si-Based Anodes: Polyacrylic Acid,” *ACS Appl Mater Interfaces*, vol. 2, no. 11, pp. 3004–3010, Nov. 2010, doi: 10.1021/am100871y.
 48. K. Ui, S. Kikuchi, F. Mikami, Y. Kadoma, and N. Kumagai, “Improvement of electrochemical characteristics of natural graphite negative electrode coated with polyacrylic acid in pure propylene carbonate electrolyte,” *J Power Sources*, vol. 173, no. 1, pp. 518–521, Nov. 2007, doi: 10.1016/j.jpowsour.2007.04.078.
 49. Z. P. Cai, Y. Liang, W. S. Li, L. D. Xing, and Y. H. Liao, “Preparation and performances of LiFePO_4 cathode in aqueous solvent with polyacrylic acid as a binder,” *J Power Sources*, vol. 189, no. 1, pp. 547–551, Apr. 2009, doi: 10.1016/j.jpowsour.2008.10.040.
 50. A. Mathew, W. van Ekeren, R. Andersson, M. J. Lacey, S. K. Heiskanen, R. Younesi, and D. Brandell, “Limitations of Polyacrylic Acid Binders When Employed in Thick LNMO Li-ion Battery Electrodes,” *J Electrochem Soc*, vol. 171, no. 2, p. 020531, Feb. 2024, doi: 10.1149/1945-7111/ad242b.
 51. L. Chai, Q. Qu, L. Zhang, M. Shen, L. Zhang, and H. Zheng, “Chitosan, a new and environmental benign electrode binder for use with graphite anode in lithium-ion batteries,” *Electrochim Acta*, vol. 105, pp. 378–383, Aug. 2013, doi: 10.1016/j.electacta.2013.05.009.
 52. N. Hamzelui, M. Linhorst, G. M. Nyenhuis, L. Haneke, G. G. Eshetu, T. Placke, M. Winter, B. M. Moerschbacher, and E. Figgemeier, “Chitosan as Enabling Polymeric Binder Material for Silicon-Graphite-Based Anodes in Lithium-Ion Batteries,” *Energy Technology*, vol. 11, no. 3, Mar. 2023, doi: 10.1002/ente.202201239.
 53. D. A. Gopakumar, A. R. Pai, D. Pasquini, L. Shao-Yuan (Ben), A. K. H.P.S., and S. Thomas, “Nanomaterials—State of Art, New Challenges, and Opportunities,” in *Nanoscale Materials in Water Purification*, Elsevier, 2019, pp. 1–24. doi: 10.1016/B978-0-12-813926-4.00001-X.

54. S. Cord-Landwehr and B. M. Moerschbacher, "Deciphering the ChitoCode: fungal chitins and chitosans as functional biopolymers," *Fungal Biol Biotechnol*, vol. 8, no. 1, p. 19, Dec. 2021, doi: 10.1186/s40694-021-00127-2.
55. L. Yue, L. Zhang, and H. Zhong, "Carboxymethyl chitosan: A new water soluble binder for Si anode of Li-ion batteries," *J Power Sources*, vol. 247, pp. 327–331, Feb. 2014, doi: 10.1016/j.jpowsour.2013.08.073.
56. K. Prasanna, T. Subburaj, Y. N. Jo, W. J. Lee, and C. W. Lee, "Environment-Friendly Cathodes Using Biopolymer Chitosan with Enhanced Electrochemical Behavior for Use in Lithium Ion Batteries," *ACS Appl Mater Interfaces*, vol. 7, no. 15, pp. 7884–7890, Apr. 2015, doi: 10.1021/am5084094.
57. J. Cui, Z. Yu, and D. Lau, "Effect of Acetyl Group on Mechanical Properties of Chitin/Chitosan Nanocrystal: A Molecular Dynamics Study," *Int J Mol Sci*, vol. 17, no. 1, p. 61, Jan. 2016, doi: 10.3390/ijms17010061.
58. J. Song, M. Zhou, R. Yi, T. Xu, M. L. Gordin, D. Tang, Z. Yu, M. Regula, and D. Wang, "Interpenetrated Gel Polymer Binder for High-Performance Silicon Anodes in Lithium-ion Batteries," *Adv Funct Mater*, vol. 24, no. 37, pp. 5904–5910, Oct. 2014, doi: 10.1002/adfm.201401269.
59. S. Deng, H. Wang, H. Liu, J. Liu, and H. Yan, "Research Progress in Improving the Rate Performance of LiFePO₄ Cathode Materials," *Nanomicro Lett*, vol. 6, no. 3, pp. 209–226, Jul. 2014, doi: 10.1007/BF03353785.
60. S. Zhang, S. Ren, D. Han, M. Xiao, S. Wang, and Y. Meng, "Aqueous sodium alginate as binder: Dramatically improving the performance of dilithium terephthalate-based organic lithium ion batteries," *J Power Sources*, vol. 438, p. 227007, Oct. 2019, doi: 10.1016/j.jpowsour.2019.227007.
61. J. Li, Y. Zhao, N. Wang, Y. Ding, and L. Guan, "Enhanced performance of a MnO₂–graphene sheet cathode for lithium ion batteries using sodium alginate as a binder," *J Mater Chem*, vol. 22, no. 26, p. 13002, 2012, doi: 10.1039/c2jm31583a
62. J. Xu, S. L. Chou, Q. F. Gu, H. K. Liu, and S. X. Dou, "The effect of different binders on electrochemical properties of LiNi_{1/3}Mn_{1/3}Co_{1/3}O₂ cathode material in lithium ion batteries," *J Power Sources*, vol. 225, pp. 172–178, Mar. 2013, doi: 10.1016/J.JPOWSOUR.2012.10.033.
63. L. Zhang, L. Zhang, L. Chai, P. Xue, W. Hao, and H. Zheng, "A coordinatively cross-linked polymeric network as a functional binder for high-performance silicon submicro-particle anodes in lithium-ion batteries," *J. Mater. Chem. A*, vol. 2, no. 44, pp. 19036–19045, Sep. 2014, doi: 10.1039/C4TA04320K.
64. Liu, Q. Zhang, Z.-Y. Wu, J.-H. Wu, J.-T. Li, L. Huang, and S.-G. Sun, "A high-performance alginate hydrogel binder for the Si/C anode of a Li-ion battery," *Chemical Communications*, vol. 50, no. 48, p. 6386, 2014, doi: 10.1039/c4cc00081a.
65. M. H. Ryou, J. Kim, I. Lee, S. Kim, Y. K. Jeong, S. Hong, J. H. Ryu, T. S. Kim, J. K. Park, H. Lee, and J. W. Choi, "Mussel-Inspired Adhesive Binders for High-Performance Silicon Nanoparticle Anodes in Lithium-Ion Batteries," *Advanced Materials*, vol. 25, no. 11, pp. 1571–1576, Mar. 2013, doi: 10.1002/adma.201203981.
66. J. Yoon, D. X. Oh, C. Jo, J. Lee, and D. S. Hwang, "Improvement of desolvation and resilience of alginate binders for Si-based anodes in a lithium ion battery by calcium-mediated cross-linking," *Phys. Chem. Chem. Phys.*, vol. 16, no. 46, pp. 25628–25635, 2014, doi: 10.1039/C4CP03499F.

Appendix

Chapter – 5 Optimization of nickel extraction from nickel sulfide ore

Table S1: EDS Point ID spectrum of roasted ore with no additive

Spectrum Label	O	Mg	Al	Si	S	Ca	Fe	Ni	Cu	Total
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Spectrum 1	58.0 5	6.26	0.62	4.04	3.82	0.24	24.88	0.55	1.56	100
Spectrum 2	67.2 9	8.77	0.33	10.48	2.85	3.77	4.61	1.17	0.73	100
Spectrum 3	72.1 8	9.07	1.67	5.38	7.39	0.65	1.44	0.44	1.78	100
Spectrum 4	67.2 2	9.3	0.52	7.19	10.88	0.14	3.23	0.1	1.41	100
Spectrum 5	65.8 4	1.61	0.2	0.75	12.96	0.18	2.2	12.85	3.4	100
Spectrum 6	54.1 3	9.15	0.26	21.33	0.73	12.32	1.72	0.11	0.24	100
Spectrum 7	68.1 1	7.16	0.08	12.91	1	7.67	2.8	0.16	0.11	100
Spectrum 8	65.1 6	0.57	0.21	0.3	0.88	0.05	32.19	0.08	0.57	100
Spectrum 9	60.5 8	1.11	0.4	0.52	1.69	0.11	34.66	0.35	0.58	100
Spectrum 10	52.3 6	4.19	0.42	2.98	4.87	0.34	20.3	14.33	0.22	100
Spectrum 11	37.3 2	2.9	0.57	2.17	6.13	0	27.81	22.48	0.61	100
Spectrum 12	62.8	1.09	0.31	0.42	5.91	0.11	18.7	9.96	0.7	100

Table S2: EDS Point ID spectrum of roasted ore with sodium sulfate additive

Spectrum Label	O	Na	Mg	Al	Si	S	Ca	Fe	Ni	Cu	Total
Spectrum 1	66.69	0.34	15.05	0.79	9.66	4.65	0.04	1.55	0.24	0.98	100
Spectrum 2	69.16	0.21	6.53	0.61	3.27	5.24	0.25	12.54	1.2	1	100
Spectrum 3	70.25	6.67	0.72	0.87	12.79	5.08	0.43	2.51	0.1	0.58	100
Spectrum 4	72	9.73	1.53	0.49	8.66	5.78	0.23	0.98	0.37	0.24	100
Spectrum 5	70.57	0.49	5.87	0.49	6.64	7.64	0.2	2.04	0.13	5.93	100

Spectrum 6	67.52	0.47	1.71	0.2	0.99	8.31	0.07	10.17	0.14	10.41	100
Spectrum 7	58.28	5.45	3.89	0.44	2.6	3.61	0.13	24.34	0.26	0.98	100
Spectrum 8	53.62	1.65	1.73	0.26	0.94	1.71	0.13	39.01	0.38	0.56	100
Spectrum 9	69.57	1.35	2.87	0.22	1.53	7.29	0.05	8.39	0.15	8.57	100
Spectrum 10	25.15	2.22	1.16	0.39	2.35	14	0.11	53.57	0.91	0.14	100
Spectrum 11	71.25	1.15	6.66	0.56	3.53	5.13	0.13	7.98	1.74	1.87	100
Spectrum 12	19.79	1.42	1.98	0.52	1.09	15.49	0.3	27.65	31.71	0.05	100
Spectrum 13	44.97	4.09	3.33	0.36	2.28	8.77	2.01	23.41	9.75	1.02	100
Spectrum 14	75.8	0.49	1.29	0.13	0.83	8.98	0.1	1.79	9.92	0.66	100
Spectrum 15	39.95	7.36	3.18	0.33	1.81	5.54	0.28	40.14	0.61	0.8	100
Spectrum 16	54.61	0.94	2.34	0.3	1.35	4.15	0.07	30.78	5.33	0.11	100
Spectrum 17	60.72	6.73	2.36	0.28	1.67	7.25	0.06	13.28	7.09	0.56	100
Spectrum 18	78.35	0.66	1.85	0.21	0.71	10.03	7.4	0.52	0.07	0.2	100
Spectrum 19	73.57	0.8	8.33	0.9	5.99	5.94	0.24	2.73	0.42	1.08	100
Spectrum 20	33.24	6.42	3.3	0.95	2.55	11.35	2.21	34.9	3.59	1.49	100
Spectrum 21	69.68	0.78	12.22	3.39	7.13	4.3	0.03	1.45	0.16	0.86	100
Spectrum 22	60.46	27.89	0.18	0.02	0.05	10.87	0.04	0.16	0.05	0.28	100
Spectrum 23	60.31	20.83	2.28	0.31	0.3	13.4	0.08	1.9	0.32	0.29	100

Table S3: EDS Point ID spectrum of roasted ore with dual salt sulfate additive

Spectrum Label	O	Na	Mg	Al	Si	S	K	Ca	Fe	Ni	Cu	Total
Spectrum 2	72.22	0.05	13.17	0.93	8.87	3.13	0.1	0	1.16	0.08	0.3	100
Spectrum 3	66.65	0.05	13.9	4.35	9.5	2.84	0.18	0.04	1.88	0.01	0.58	100
Spectrum 4	74.22	0.36	7.6	2.6	6.3	5.36	0.05	0.05	1.91	0.11	1.43	100
Spectrum 5	68.3	0.33	7.32	1	12.83	1.39	0.1	4.93	3.04	0.49	0.26	100
Spectrum 6	62.09	23.47	0.62	0.05	0.05	11.75	0.59	0.03	0.76	0.22	0.37	100
Spectrum 7	71.73	0.48	10.04	1.26	4.62	6.29	0.3	0.08	3.19	0.17	1.82	100
Spectrum 8	69.66	0.26	2.44	0.18	1.29	8.56	0.11	0.14	5.61	0.2	11.55	100
Spectrum 9	53.45	0.85	0.62	0.23	0.24	1.72	0.09	0.03	41.93	0.56	0.26	100
Spectrum 10	69.19	0	2.4	0.33	1.4	0.92	0.05	0.05	24.73	0.64	0.28	100
Spectrum 11	28.24	1.8	2.89	0.61	1.44	4.57	1.84	0.25	57.07	0.72	0.56	100
Spectrum 12	53.49	2.62	2.27	0.36	1.55	16.43	0.18	0.31	3.29	19.32	0.19	100
Spectrum 13	68.56	1.02	4.09	0.42	2.22	7.49	0.12	0.07	7.15	7.65	1.23	100
Spectrum 14	30.44	1.58	3.25	0.33	1.56	8	0.9	0.39	43.14	9.48	0.91	100
Spectrum 15	67.81	0.25	8.32	0.55	3.62	9.89	0.11	0.18	6.63	1.23	1.4	100
Spectrum 16	58.23	0.91	2.35	0.25	0.79	10.62	0.96	0.03	12.1	12.37	1.37	100
Spectrum 17	71.81	3.7	4.44	0.24	0.99	11.25	0.22	1.32	5.1	0.7	0.22	100
Spectrum 18	70.63	1.52	3.13	0.35	1.81	12.42	0.37	7.19	1.97	0.31	0.31	100
Spectrum 19	46.24	4.01	3.07	0.67	2.18	14.52	0.7	0.18	21.09	6.57	0.77	100
Spectrum 20	59.82	20.11	0.91	0.1	0.05	15.74	0.47	0.05	1.98	0.33	0.43	100
Spectrum 21	69.19	0.32	1.68	0.31	1	12.63	10.46	0.2	3.81	0.24	0.14	100
Spectrum 22	75.78	0.67	4.45	0.18	0.37	9.79	6.01	0.16	1.26	0.92	0.42	100