Faculty of Electrical Engineering
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Doctoral Thesis

Lithium-Ion Battery Recycling Pre-Treatment: A Techno-Environmental-Economic Perspective

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Honour Statement

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Last but not least, *I want to thank me.*

Abstract

The rapid expansion of lithium-ion battery (LIB) usage across consumer electronics, electric vehicles, and energy storage systems intensifies the demand for sustainable end-of-life management. Driven by the global shift towards decarbonisation and electrification, the production and deployment of LIBs have grown exponentially, raising urgent concerns regarding the environmental and economic consequences of improper disposal. The composition of LIBs is complex and contains potentially hazardous components, including flammable electrolytes and critical raw materials such as cobalt, nickel, manganese, and lithium, the recovery of which is essential to reduce environmental burdens and limit dependence on primary resources. Therefore, developing effective recycling strategies is key to resource conservation, environmental protection, and establishing a circular economy.

While downstream recycling techniques, such as pyrometallurgy, hydrometallurgy, and direct recycling, have received significant attention in research, their performance is strongly influenced by the quality of input materials, which is shaped mainly during the pre-treatment phase. Despite the recognised importance of pre-treatment, there is still limited comprehensive insight into this stage, particularly regarding safety, cost-effectiveness, and environmental impacts. Further optimisation of operations such as sorting, discharging, disassembly, and material separation is recommended to ensure high-quality outputs and safe processing conditions, thereby enabling efficient downstream recovery and minimising material losses.

This dissertation addresses these challenges by evaluating and improving pre-treatment strategies through experimental research and sustainability assessment. Three key areas are examined: (i) the safe and efficient electrochemical discharging of LIBs using aqueous solutions; (ii) the enhancement of black mass quality via the optimisation of recycling pre-treatment steps; and (iii) the environmental and economic performance of selected pre-treatment approaches, with a regional focus on the Czech Republic. Methodologies include experimental trials, life cycle assessment, and cost-benefit analysis. The expected contribution of this work lies in delivering a comprehensive, process-level understanding of pre-treatment and its role in improving the efficiency, safety, and sustainability of LIB recycling. This research supports regulatory compliance, fosters circularity, and informs the development of scalable recycling infrastructure aligned with EU goals.

Keywords: Lithium-Ion Battery Recycling, Recycling Pre-Treatment, Electrochemical Discharging, Black Mass Quality, Sustainability, Resource Recovery, Circular Economy.

Anotace (in Czech)

Rychlý rozvoj využívání lithium-iontových baterií napříč oblastmi spotřební elektroniky, elektromobility a energetických úložišť výrazně zvyšuje potřebu jejich udržitelného nakládání na konci životnosti. V důsledku globálního posunu směrem k dekarbonizaci a elektrifikaci dochází k prudkému nárůstu výroby a používání lithium-iontových baterií, což vyvolává obavy ohledně environmentálních a ekonomických dopadů z jejich nesprávné likvidace. Složení lithium-iontových baterií je komplexní a obsahuje i potenciálně nebezpečné složky, včetně hořlavých elektrolytů a kritických surovin, jako jsou kobalt, nikl, mangan a lithium. Jejich efektivní zpětné získávání je klíčové pro snížení ekologické zátěže a omezení závislosti na primárních zdrojích. Proto je nezbytný rozvoj účinných recyklačních strategií, které umožní ochranu životního prostředí, zachování zdrojů a přechod k cirkulárnímu bateriovému hospodářství.

Ačkoli byla metodám následného zpracování, jako je pyrometalurgie, hydrometalurgie či přímá recyklace, věnována značná pozornost, jejich efektivita je výrazně ovlivněna kvalitou vstupního materiálu, která je formována především během recyklační předúpravy. Navzdory důležitosti této fáze zůstává její komplexní pochopení stále omezené, zejména pokud jde o otázky bezpečnosti, ekonomické efektivity a environmentálních dopadů. Pro dosažení kvalitních výstupů a zajištění bezpečných pracovních podmínek je proto doporučována další optimalizace jednotlivých kroků, jako je třídění, vybíjení, demontáž a separace jednotlivých materiálových složek.

Tato disertační práce se zaměřuje na výzkum a zefektivnění procesů recyklační předúpravy prostřednictvím experimentálního ověřování a hodnocení udržitelnosti. Řešené jsou tři klíčové oblasti: (i) bezpečné a efektivní elektrochemické vybíjení lithium-iontových baterií za použití vodných roztoků, (ii) zlepšení kvality výstupní černé hmoty s pomocí optimalizovaných kroků recyklační předúpravy a (iii) environmentální a ekonomické hodnocení vybraných přístupů předúpravy se zohledněním podmínek České republiky. Použité metody zahrnují laboratorní experimenty, analýzu životního cyklu a ekonomické zhodnocení nákladů a přínosů. Přínos práce spočívá v detailním porozumění roli recyklační předúpravy při zvyšování efektivity, bezpečnosti a udržitelnosti procesů zpracování odpadních lithium-iontových baterií. Výsledky podporují směr platné bateriové legislativy, přispívají k posílení cirkulárního hospodářství a poskytují podklady pro rozvoj škálovatelné recyklační infrastruktury v souladu s cíli EU.

Klíčová slova: recyklace lithium-iontových baterií, recyklační předúprava, elektrochemické vybíjení, kvalita černé hmoty, udržitelnost, získávání zdrojů, cirkulární ekonomika.

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Abbreviations

BESS	Battery Energy Storage System
BEV	Battery Electric Vehicle
BM	Black Mass
CEC	Consumer Electronics Cell
DEC	Diethyl Carbonate
DMC	Dimethyl Carbonate
EDS	Energy Dispersive Spectrometry
EMC	Ethyl Methyl Carbonate
EOL	End-of-Life
EU	European Union
EV	Electric Vehicle
GC-MS	Gas Chromatography-Mass Spectrometry
GS	Google Scholar
HEV	Hybrid Electric Vehicle
HPLC	High-Performance Liquid Chromatography
IRS	Infrared Spectroscopy
ICP-OES	Inductively Coupled Plasma-Optical Emission Spectrometry
IF	Impact Factor
LIB	Lithium-Ion Battery
LCA	Life Cycle Assessment
LCO	Lithium Cobalt Oxide
LFP	Lithium Iron Phosphate
LMO	Lithium Manganese Oxide
NCA	Lithium Nickel Cobalt Aluminium Oxide
NMC	Nickel Manganese Cobalt Oxide
NMP	N-Methyl Pyrrolidone
PE	Polyethylene
PHEV	Plug-In Hybrid Electric Vehicle
PP	Polypropylene
PVDF	Polyvinylidene Fluoride
SOC	State-of-Charge
WoS	Web of Science
XRD	X-Ray Diffraction
XRF	X-Ray Fluorescence

Introduction

Lithium-ion batteries (LIBs) have significantly reshaped the energy storage landscape, becoming indispensable for modern society [1]–[3]. Their high energy density, relatively long cycle life, and decreasing costs have made them the dominant power source for a vast array of applications [4], ranging from portable consumer electronics (CECs) like smartphones and laptops to large-scale systems such as electric vehicles (EVs) and stationary battery energy storage systems (BESSs) [5]–[8]. The global transition towards electrification, especially in transportation, and the integration of intermittent renewable energy sources into power grids drive exponential growth in LIB production and deployment [9]–[11]. This surge represents a global commitment to cleaner energy technologies, but it also presents a critical challenge of sustainably managing the increasing volume of end-of-life (EOL) LIBs [12]–[15].

With the growing number of EOL LIBs, developing and implementing efficient, safe, and economically viable recycling technologies has become essential. Improper handling or disposal of spent batteries can release hazardous substances such as heavy metals or flammable electrolytes, posing serious environmental and safety risks [16]–[18]. At the same time, LIBs contain valuable and non-renewable materials like lithium, cobalt, nickel, manganese, copper, and aluminium [4]. Their recovery can significantly reduce the need for primary raw material extraction, which is often associated with geopolitical instability, environmental degradation, and human rights concerns [19], [20]. From both ecological and economic standpoints, advanced recycling strategies support resource conservation and waste reduction, enhance supply chain resilience, and contribute to developing a circular battery economy, which is essential for a sustainable energy transition [21]–[24].

To address these issues, regulatory frameworks are tightening. A key example is the Battery Regulation (EU 2023/1542) [25], which sets mandatory targets across the battery lifecycle, from raw material sourcing to EOL management. For LIBs, it mandates minimum recovery rates for critical materials, such as 50% lithium by 2027, 80% by 2031, and 90% cobalt, copper, lead, and nickel by 2027, increasing to 95% by 2031. The new Battery Regulation also sets overall recycling efficiency targets of 65% by 2025 and 70% by 2030, as illustrated in Fig. 1 [25]. These regulations reflect the growing societal and political will to boost recycling efficiency and drive the urgent need for technological innovation in battery recycling, especially in the pre-treatment phase.

While metallurgical processes, such as pyrometallurgy, hydrometallurgy, or direct recycling, are essential for the recovery of purified materials, their efficiency depends heavily on the quality of the input, which is determined during pre-treatment [17], [26]–[29].

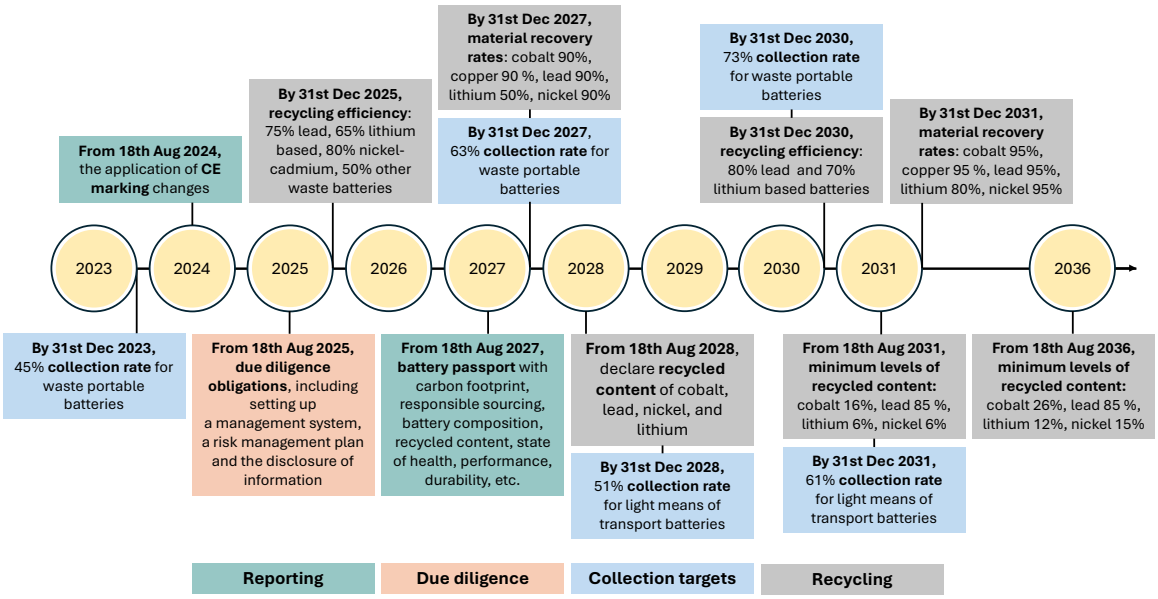

Figure 1: Timeline of key milestones from the Battery Regulation (EU 2023/1542), focusing on circular economy aspects [25].

This preparatory stage encompasses various physical and chemical operations that make EOL batteries safer to handle and prepare for downstream recovery [30]–[32]. Its primary function is to convert complex and potentially hazardous battery waste into more uniform, safer, and material-enriched input streams suitable for efficient downstream processing [27], [30]. Pre-treatment typically includes battery sorting, discharging to eliminate residual charge and prevent short circuits, disassembly to module or cell level, and mechanical processing (e.g., shredding, crushing) as illustrated in Fig. 2 [30], [33], [34]. This is followed by separation processes, such as sieving, magnetic separation, and density-based sorting, to isolate components like steel casings, aluminium and copper foils, plastics, and electrode powders [35]–[37]. Optional steps like thermal or chemical treatments may be applied to remove volatile compounds, increase purity, and improve metal recovery [30], [34]. A crucial product of pre-treatment is a fine powder referred to as "black mass" (BM), which consists mainly of active electrode materials, including lithium, cobalt, nickel, manganese, and graphite, along with residual electrolyte, binders, and conductive additives [30]–[33]. The composition and quality of BM depend on the efficiency of upstream operations and significantly impact the cost and performance of subsequent recycling [38].

Designing effective recycling pre-treatment processes involves balancing safety, economic viability, and environmental sustainability. Even in their EOL state, LIBs may contain residual charge and chemically reactive materials, posing explosion risks [18], [39]–[41]. While stable under normal conditions, mechanical damage or poor handling can release flammable electrolytes, heavy metals, or toxic gases [16], [39], [42]. Processing must occur

in controlled environments to mitigate these risks, with proper ventilation, personal protective equipment, and safe discharging protocols [42], [43]. Economics also plays a key role: pre-treatment involves capital expenditure and operation costs (e.g., energy, labour, maintenance, waste management) [44]. Material loss during pre-treatment can reduce economic returns, so maximising recovery, minimising energy use, and reducing waste are essential for profitability [44], [45]. Enhancing overall economic performance also encourages further investment and scale-up. Sustainable pre-treatment aims to increase the yield of secondary raw materials, reduce environmental harm, and support a circular battery value chain [29], [45]–[47].

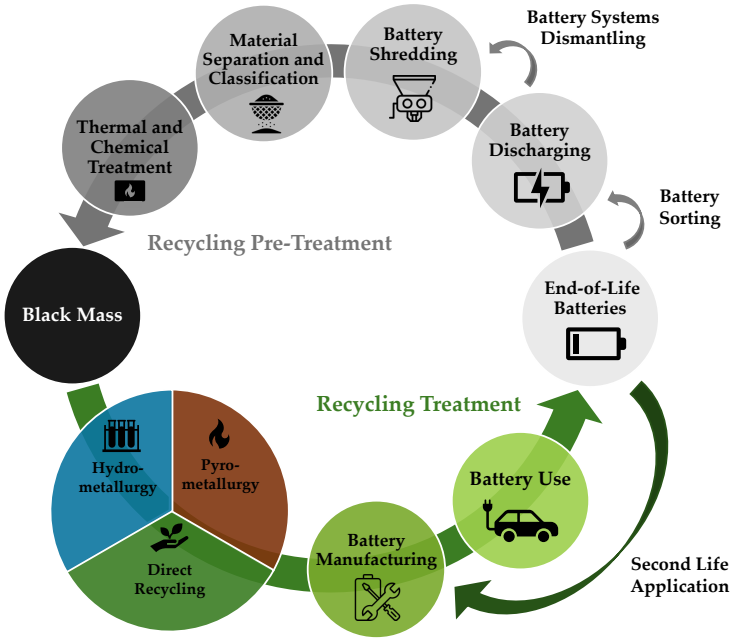

Figure 2: The scope and sequence of lithium-ion battery end-of-life treatment, including recycling pre-treatment, recycling processes, and second life application.

While recovery processes like pyrometallurgy and hydrometallurgy have garnered significant attention, the pre-treatment stage of LIB recycling remains comparatively underexplored. There is limited experimental data on safe and scalable sorting [7], [48], [49], disassembly [48], [50], [51], discharging [52]–[55], mechanical processing [56], [57], separation [32], and post-treatment methods, particularly regarding their environmental and economic impacts [29], [58]. This dissertation addresses these gaps by focusing on pre-treatment, its influence on the quality of recovered materials, and the feasibility of different recycling approaches. The main objective is identifying and assessing strategies that improve safety and material recovery efficiency while minimising the associated environmental and economic burdens. Accordingly, this work aims to support preparing high-quality intermediate materials tailored for subsequent recycling processes, including pyrometallurgy, hydrometallurgy, and direct recycling.

The research addresses three main thematic areas, each reflecting a critical aspect of the LIB recycling pre-treatment process. The overarching objective is to comprehensively understand and assess how pre-treatment can improve process safety, material recovery, and sustainability in LIB recycling. Each thematic area is structured around a specific research objective:

- **Electrochemical discharging of batteries**: The first objective is to evaluate the feasibility and effectiveness of aqueous discharging methods across different battery chemistries. This part investigates the effect of aqueous salt solutions on the discharging efficiency, aiming to reduce environmental risks (e.g., electrolyte leakage, wastewater contamination) and minimise material losses. Additionally, the goal is to assess operational challenges and explore hazard mitigation strategies, providing a balanced view of industrial applicability and process sustainability. Special attention is given to the potential of non-invasive discharging concepts and their comparability to conventional methods regarding safety, effectiveness, and compatibility with downstream recycling processes.

- **Enhancement of BM quality through pre-treatment steps**: The second objective is to identify and experimentally validate improvements in pre-treatment that enhance the quality and consistency of BM. This section focuses on improving key processing steps, including battery sorting, disassembly, and environmentally sustainable separation techniques. Furthermore, this part aims to refine thermal conditioning to increase the purity and homogeneity of recovered fractions.

- **Evaluation of environmental and economic performance of recycling pre-treatment strategies**: The third objective is to assess the sustainability of selected pre-treatment pathways using life cycle assessment and techno-economic analysis. This part intends to quantify material and energy flows, identify major environmental impact contributors, and evaluate region-specific conditions relevant to Central European recycling infrastructure, particularly in the Czech Republic.

The expected contribution of this work is a detailed, process-level evaluation of LIB recycling pre-treatment strategies, grounded in experiments and sustainability metrics. This dissertation aims to support the development of safer, more efficient, and scalable recycling solutions within a circular battery economy by addressing the defined objectives.

Chapter 1

Study of Electrochemical Lithium-Ion Battery Discharging

The safe and efficient EOL management of LIBs presents a significant challenge for sustainable battery recycling. A critical aspect of this process is removing residual charge during the recycling pre-treatment, which helps mitigate the dangers of short circuits, thermal runaway, and explosion hazards during transport, storage, and processing [59]–[61]. To minimise these risks and ensure the safety and efficiency of subsequent recycling steps, including mechanical processing such as crushing and shredding [62], [63], generally speaking, the LIBs should ideally undergo a deep discharge to below 2.5 V per cell, with a preferred safety threshold under 1 V. This process induces irreversible electrochemical degradation in the cathode active material, leading to structural changes that prevent the battery from regaining charge and thereby further enhancing safety [64], [65].

Various strategies have been explored for the safe and controlled discharge of LIBs. Active dischargers, typically used for EV batteries, allow for energy recovery and precise control of the discharge process. On the other hand, passive methods, such as freezing batteries with liquid nitrogen [59], [66] or processing them in inert atmospheres [67]–[69], have been investigated to reduce thermal and electrical hazards. However, economic and logistical constraints hinder the widespread implementation of these methods [60].

Electrochemical discharge in aqueous salt solutions is one of the most widely applied methods for CECs due to its relative simplicity, scalability, and effectiveness in reducing residual voltage before mechanical processing [55], [70]–[74]. Although this method has been studied for over 20 years [75] and is generally regarded as effective, recent research has identified several challenges, such as incomplete discharge, voltage rebound, and corrosion induced by discharging media, leading to contamination [53], [54], [73], [76]. Additionally, inconsistencies in discharge efficiency across different solutions raise concerns about the reliability of this approach, especially when considering the increasing variety of waste battery compositions, both current and future [52], [59].

Addressing the limitations of electrochemical discharge methods, such as incomplete discharge, voltage rebound, and contamination, is essential for advancing LIB recycling. Therefore, ongoing research efforts focus on improving the efficiency and environmental

compatibility of discharge processes for CECs. This involves a twofold approach: optimising existing electrochemical techniques to overcome known drawbacks and exploring alternative discharge strategies that offer similar performance without these disadvantages. These innovations aim to deliver safe, scalable, and sustainable solutions for CECs. Integrating such improved discharge methods into industrial recycling workflows is crucial for promoting circular economy principles, and the development of standardised, scalable procedures will support greater resource efficiency and safety within the transition to a low-carbon energy system.

1.1 Author's Contribution to This Research Field

Electrochemical discharging, while widely used, presents several challenges, particularly regarding its environmental impact and economic efficiency. This work addresses these issues by studying discharging processes to improve discharge efficiency and sustainability. It examines the environmental and safety concerns associated with the electrochemical discharging techniques and the economic implications of material losses. It highlights the need for better waste management strategies and more cost-effective approaches to LIB recycling.

The contributing study [C1] investigated the process efficiency and environmental perspective of sodium chloride (NaCl) solution discharging. Specifically, the analysis involved an efficiency evaluation for varying solution concentrations (5 wt.% and 10 wt.%) for cylindrical LIB cells in both nickel-manganese-cobalt oxide (NMC) and lithium-iron-phosphate (LFP) chemistries, with capacities ranging from 1.5 to 5 Ah, in their original shipped or fully charged state. The study assessed the efficacy of NaCl solutions in achieving discharge to a safe voltage limit of 0.75 V within a 24-hour timeframe, proving this process as a suitable pre-treatment step for ensuring the safety of subsequent recycling stages. All tested cells were discharged to the target voltage limit. However, it was observed that the discharge was primarily driven by cell reaction-based contact damage, leading to the battery contents' leakage into the solution, as illustrated in Fig. 1.1.

A detailed characterisation of the resultant waste solutions revealed the presence of hazardous volatile organic compounds, including carbonic acid esters, methyl salicylate, and propanoic acid esters, identified via Gas Chromatography-Mass Spectrometry (GC-MS). Furthermore, Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) detected a high proportion of metals within the wastewater medium, which could lead to potential economic and material losses with improper treatment. This work highlights the necessity of implementing rigorous waste management protocols in LIB recycling to mitigate the environmental risks associated with hazardous by-products. It emphasises the importance of integrating efficient discharge processes with responsible waste management. It underscores the potential for valuable metal recovery through techniques such as hydrometallurgy, contributing to a more sustainable closed-loop recycling process.

In work [C2], which builds upon previous research [C1], the further challenges associated with the electrochemical discharging of other cylindrical LIB cells were studied in detail.

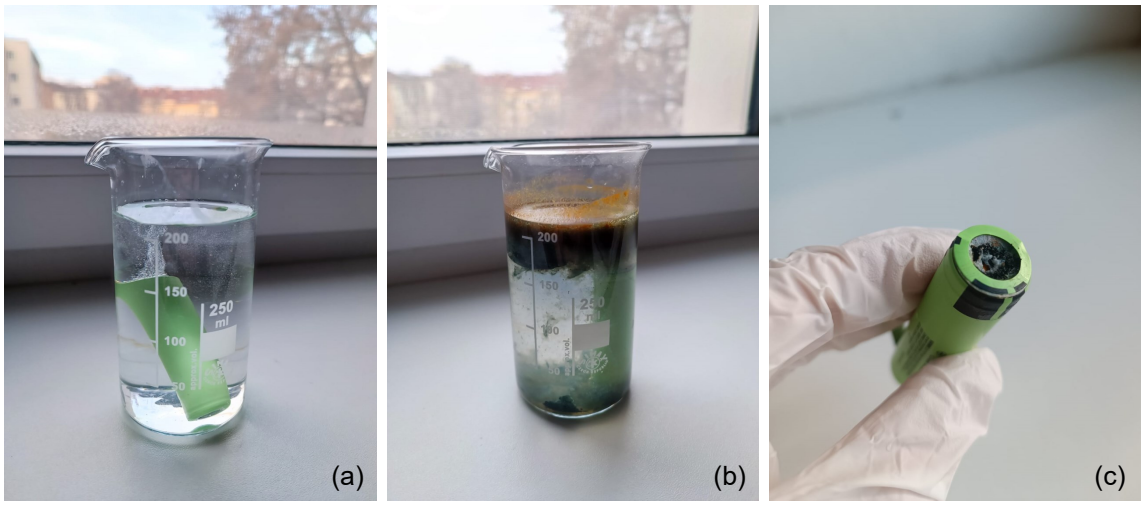

Figure 1.1: Electrochemical discharging of cylindrical LIB cells. Illustration of (a) initial discharging reactions; (b) solution after 14-hour discharging procedure, with reacted and released cell contents; (c) visible damage to the battery contact caused by the discharging process. Further details are provided in works [C1] and [C2].

This work focused exclusively on 18650 cells in their original shipped state, covering three chemistry types: LFP, NMC, and nickel-aluminium-cobalt (NCA), which were exposed to aqueous solutions of NaCl, sodium hydroxide (NaOH), and sodium nitrate ($NaNO_3$), with concentrations ranging from 5 to 30 wt.%. The analysis involved characterising the discharging process and examining the quantity and composition of the mass released from these cells due to cell contact damage. Discharging was achieved for all battery types to a voltage limit below 1 V, as shown in Fig. 1.2. However, for all treated cells, discharging, which led to the leakage of battery contents into the discharging solutions, occurred again.

Notably, NCA cells showed the highest percentage of mass release, with the most significant battery content release occurring when using 30 wt.% NaCl solutions. Thus, for this scenario, potential economic losses were estimated based on the released quantities of nickel, copper, aluminium, iron, and manganese. The results indicated total metal material cost losses ranging from \$190 to \$1,560 per tonne of treated NCA LIB cells. This result underscores the importance of implementing effective waste management strategies to reduce material losses and enhance the recovery of valuable metals, thereby supporting a more sustainable approach to LIB recycling.

The following study [C3] expanded on previous research on electrochemical discharging [C1, C2] by focusing on the safety risks and environmental hazards of the wastewater generated during the EOL treatment processes. Previous studies primarily examined cylindrical cells, where different solutions were used for electrochemical discharging to achieve a safe voltage threshold. However, these studies did not fully address the toxic components in the wastewater, which included organic solvents from the electrolyte, thereby providing valuable insights into the environmental risks associated with their recycling.

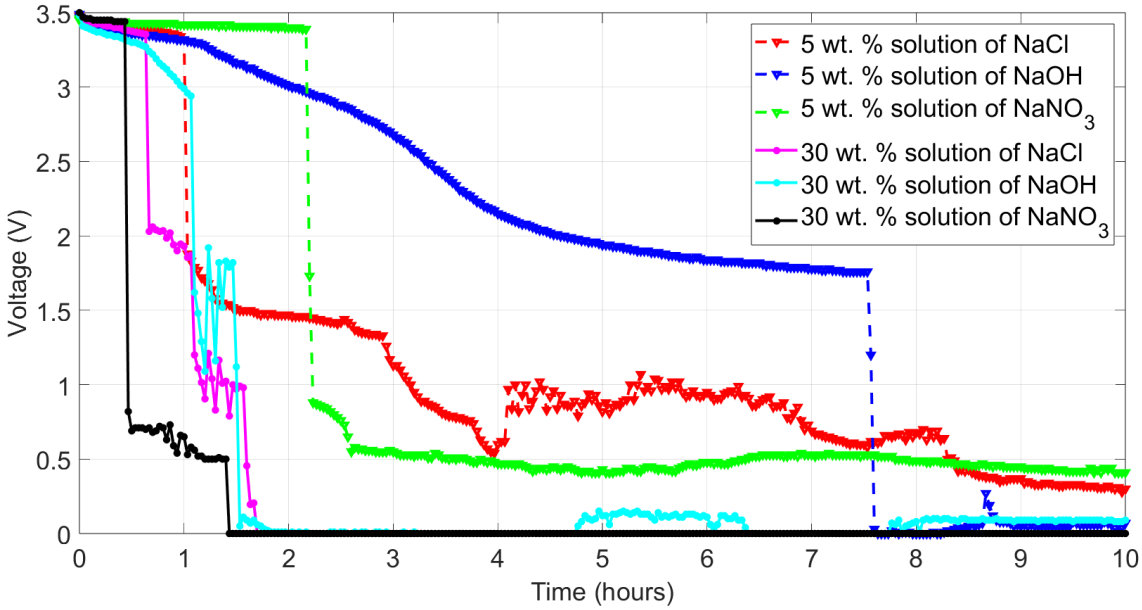

Figure 1.2: Example of discharging profiles measured for lithium nickel-cobalt-aluminium oxide (NCA) battery cells, electrochemically discharged in NaCl, NaOH, and $NaNO_3$ solutions at concentrations of 5 wt.% (dashed lines) and 30 wt.% (solid lines). © 2024 IEEE. Reprinted with permission from [C2].

This work thoroughly examines the released substances, including metals and electrolyte components, by analysing the water-based crushing of deeply discharged pouch NMC cells (3.65 V, 78 Ah). The analysis revealed that while the initial contamination was substantial, particularly the electrolyte, repeated water flushing significantly reduced the pollutant concentrations, with the first flush proving most effective. This finding suggests that pre-flushing cells after discharging, before further processing (e.g., crushing or storage), could be an effective strategy for reducing the release of harmful substances. These insights emphasise that pursuing safer and more sustainable discharge methods must be a central focus of LIB recycling research.

An alternative approach to conventional electrochemical discharging was developed as part of this doctoral research. This method is based on non-electrochemical discharging principles and is described in detail in a Czech national patent [P1]. The core idea is to discharge LIB cells by generating a short-circuit current through conductive metal fragments dispersed in a cooling liquid, directly connecting the electrode terminals. This allows for rapid voltage reduction without mechanical damage to the cell casing and without releasing battery materials or electrolyte into the surrounding medium. LIB cells at approximately 30% state-of-charge (SOC) typically drop below 1 V within 30 minutes and often below 0.5 V within 15 minutes. Complete discharging (with no significant voltage rebound effect) is achieved by discharging the cells for a longer period, typically at least 15 hours for cells at 30% SOC, 5 hours for EOL cells, and 18 hours for fully charged

cells. This leads to a final stabilised voltage below 1 V after 24 hours of regeneration. The entire process occurs without thermal runaway, sparking, or internal short-circuiting. The achieved final voltage level is considered safe for subsequent recycling steps, such as crushing and shredding, and the method provides a comparable or even superior discharging performance to conventional electrochemical methods. Its non-invasive nature, safety, and scalability make it a highly promising pre-treatment method for battery recycling.

Comparative experiments using GC-MS analysis were conducted to evaluate cell integrity and the degree of contamination in the cooling and discharging media, as illustrated in Fig. 1.3.

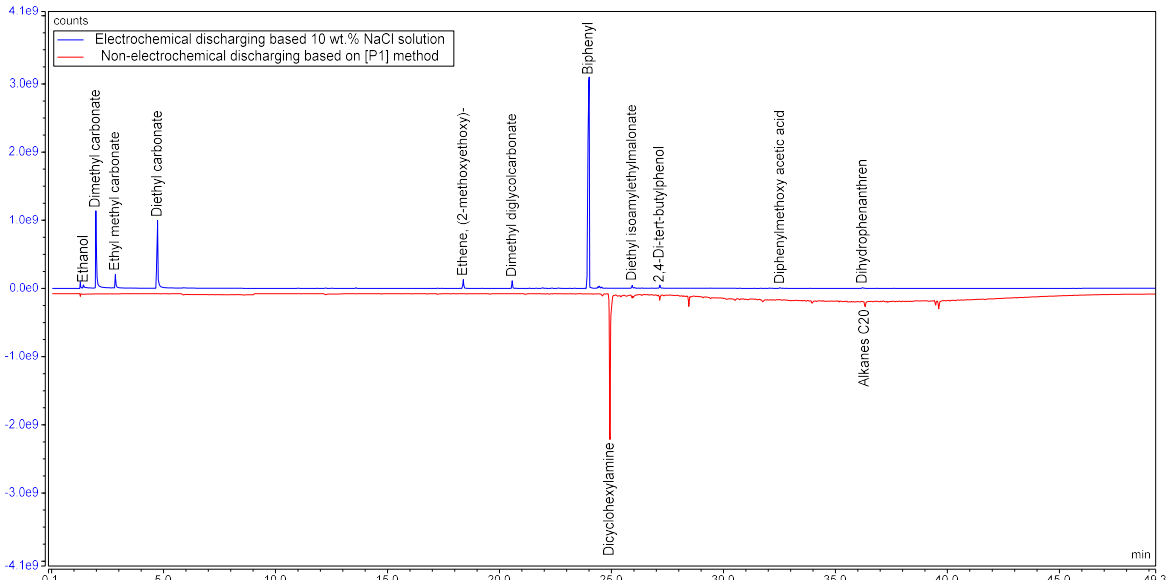

Figure 1.3: Comparison of chromatograms of discharging solution (electrochemical method) and coolant samples (non-electrochemical discharging method) from cylindrical lithium-ion battery cells (HW 22650-3M, 3.7 V, 2600 mAh, PA15).

Coolant samples, based on deionised water in High-Performance Liquid Chromatography (HPLC) gradient grade, obtained through this alternative discharging method using iron filings derived from metal machining as the short-circuiting medium, were compared to those from conventional electrochemical discharging in 10 wt.% NaCl solution. Due to the machining origin of the filings, residues of cutting oil, specifically antiwear agents such as dicyclohexylamine, and long-chain alkanes (around C20) were observed in the coolant [77], [78]. The absence of key electrolyte components such as dimethyl carbonate (DMC), ethyl methyl carbonate (EMC), and diethyl carbonate (DEC) [4], [43], [79] in the coolant confirmed that no electrolyte leakage occurred, thereby preserving cell integrity throughout the process. In contrast, these carbonate solvents were consistently detected in electrochemically discharged samples, indicating electrolyte release and potential cell rupture. Furthermore, specific degradation products, such as ethanol (a decomposition product of carbonate solvents), were exclusively detected in electrochemical discharging

solution samples [43]. The presence of ethene-(2-methoxyethoxy)-, commonly found in flux agents, was also observed [79]. The rest of the compounds, identified from biphenyl onwards, likely represent plasticisers leaching from battery casing materials [77], [79]. Importantly, despite its non-invasive nature, the non-electrochemical method achieved complete discharging within a timeframe comparable to conventional approaches. The partial or complete elimination of dicyclohexylamine and alkanes in the coolant could be improved by pre-washing the iron filings with a solvent such as propanol before the discharging procedure. This demonstrates that the approach provides superior environmental and operational safety while matching the functional performance of traditional electrochemical discharging. It therefore represents a highly promising and practical alternative for industrial battery recycling workflows.

1.2 Conclusions and Perspectives

Electrochemical discharging in aqueous media has proven effective for lowering the residual voltage of LIBs. However, the findings also highlight several environmental and cost-related concerns linked to its implementation. The release of volatile organic compounds and elevated concentrations of metals into wastewater during the discharging process raises significant concerns regarding the safe management of liquid effluents. Additionally, the influence of battery chemistry, state of charge, and solution concentration on the discharging efficiency indicates a need for further optimisation to avoid material losses and ensure consistency across different LIB types. The effectiveness of post-discharge treatments, such as water flushing, has been demonstrated in mitigating the environmental risks linked to harmful substances and thus represents a viable measure to improve process safety and reduce the overall environmental burden.

These results emphasise the importance of incorporating effective and controllable discharging strategies into the broader framework of sustainable LIB recycling. Developing discharge protocols that minimise environmental impact while maintaining material integrity for downstream recovery is essential to ensure alignment with circular economy objectives. Integrating tailored electrolyte removal approaches, improved flushing techniques, and robust waste handling procedures will be necessary to reduce secondary contamination and support the recovery of valuable components. In this context, the novel non-invasive method demonstrated here proves particularly advantageous, as it preserves cell structure and avoids electrolyte leakage, reducing the need for additional purification steps in subsequent processing.

Future work should prioritise refining discharging methodologies to enable safer, scalable, and more environmentally benign alternatives to existing practices. This includes exploring non-aqueous or solvent-free approaches and evaluating process conditions that may facilitate the selective removal of reactive components without compromising the integrity of valuable materials. Furthermore, targeted development of wastewater treatment strategies with integrated resource recovery, particularly for metals and organic compounds, may significantly improve the process's economic viability and environmental performance.

Chapter 2

Improvement Strategies for Black Mass Quality Optimisation

The quality of BM in LIB recycling is directly influenced by the efficiency of pre-treatment processes, such as sorting, discharging, dismantling, comminution, classification, separation, and additional post-processing methods, including chemical leaching or high-temperature treatment [30]. These steps determine the final composition and purity of the BM, making their optimisation crucial for enhancing metal recovery rates and achieving high-quality products [24], [27], [30], [38].

Battery sorting, based on both battery architecture and material composition, ensures that materials are processed more efficiently and that the BM has a more controlled and predictable composition [17], [30]. However, the main challenge lies in developing sorting systems that can accurately and quickly classify LIBs based on their specific chemical makeup [7], [48]. Inaccurate or poorly managed sorting can lead to contamination of the BM with unwanted materials, such as plastics or other metals, which degrade its quality and may require more complex and resource-intensive separation steps to achieve the desired purity [29], [49].

The type of discharging process used, as discussed in Chapter 1, can also influence the final composition of the BM. Electrochemical discharging methods can introduce residual salts or reaction byproducts, which may alter the chemical composition of the recovered materials [54], [73], [74]. Optimising these discharging methods is crucial to minimising such contaminants, enhancing process safety, and ensuring the purity consistency of the BM.

Dismantling, both manual and automated, represents a crucial step influencing the efficiency of subsequent treatments [27], [30]. Manual dismantling can offer a degree of precision in component separation, but is prone to human error, making it less reliable in large-scale operations [51]. On the other hand, automated dismantling systems provide higher throughput and can be calibrated in operation to optimise separation efficiency [50], [51], [80]. However, poorly executed dismantling processes may result in non-target materials remaining in the BM, which can adversely affect the overall quality of the material and reduce the effectiveness of subsequent recovery stages [48], [51].

The comminution process, which involves crushing and grinding, is critical for liberating electrode materials from current collectors and preparing the BM for subsequent recovery [30], [63]. However, excessive or improper comminution can lead to an inefficient separation of materials and result in the loss of valuable components [56]. Achieving the optimal balance between energy consumption, particle size reduction, and effective material separation is essential for maintaining the quality of the BM [56], [57], [81]. The selection of appropriate classification techniques further influences the composition of the recovered products, ensuring that valuable materials are preserved for effective recovery [29], [56], [58].

Additional post-processing methods play a vital role in refining the composition of the BM and improving the efficiency of direct metal recovery [30]. Chemical leaching selectively dissolves specific components, facilitating the separation of valuable metals while minimising impurities [34], [82], [83]. High-temperature treatments, including pyrolysis or thermal decomposition, can further aid in removing organic binders and residual electrolytes, reducing contamination and enhancing the purity of recovered metals [23], [82], [84], [85]. Integrating these methods into the recycling pre-treatment process structure is essential for achieving high recovery yields and ensuring that the extracted metals meet the required requirements for reuse in battery manufacturing [85].

Ensuring efficient pre-treatment processes in LIB recycling is essential for achieving high-quality BM and optimising subsequent metal separation [30], [85]. Careful control of each step—from sorting to additional treatments—minimises the loss of valuable materials and reduces the energy and material demands of the entire process [17], [23], [45]. A systematic approach to these technologies directly impacts recycling practices' economic and environmental sustainability, paving the way for their broader industrial application [47], [85].

2.1 Author's Contribution to This Research Field

Technological advancements play a crucial role in minimising contamination and enhancing the purity of BM, ultimately improving metal recovery efficiency in LIB recycling. This work contributes to developing optimised pre-treatment strategies, emphasising their role in refining material separation and maximising recovery yields. By integrating innovative approaches to sorting, dismantling, material characterisation, separation and additional treatments, this research advances the field toward more effective and sustainable LIB recycling processes.

Conventional four-step pre-treatment approaches are increasingly considered inadequate for addressing contemporary industrial demands. As highlighted in a comprehensive review of LIB recycling technologies [J1], the pre-treatment phase is a critical factor influencing overall process efficiency and economic viability. This review addresses a more encompassing seven-step process, encompassing battery discharging, dismantling, comminution, classification, separation, dissolution, and thermal treatment, to better align with current industrial practices. The selection of processing methods at each stage governs the yield and composition of the resulting BM, thereby exerting a cascading effect

on the efficiency of downstream recovery steps. These steps are essential, as the composition and properties of the BM greatly influence the efficiency of the subsequent metal recovery processes.

To address the need for efficient material characterisation in optimising BM quality, the [C4] study presented a methodology for LIB cells' comprehensive, rapid, cost-effective, and accurate material composition survey. This work, focusing on cylindrical (18650) cells with NCA and NMC chemistries, employed manual disassembly followed by detailed chemical analysis using Scanning Electron Microscopy (SEM) with Energy-Dispersive X-ray Spectroscopy (EDS). The presented approach enabled the precise determination of battery component proportions and the chemical composition of various cell parts, including the cathode, anode, separator, and metal casing.

The resulting step-by-step protocol facilitates the acquisition of essential chemical and material parameters, evaluates recycling potential and provides critical data for developing advanced robotics and automation in recycling processes. This outcome can lead to accelerated and more precise battery sorting, ultimately improving the quality of BM and other outputs, such as battery casings and current collectors.

Further research [C5] investigated the initial steps of the recycling pre-treatment procedure for the EOL battery electric vehicle (BEV) LIB module (29.36 V, 6.85 kWh, in deeply discharged state). This work detailed the module-to-cell disassembly process and the material analysis of its components. The methodology included a manual disassembly approach, as illustrated in Fig. 2.1, followed by Electrochemical Impedance Spectroscopy (EIS) and Cyclic Voltammetry (CV) to characterise the cell's discharge state. X-ray Fluorescence (XRF) and X-ray Diffraction (XRD) were employed to determine the material composition of the battery cell components, focusing on the proportions of valuable metals.

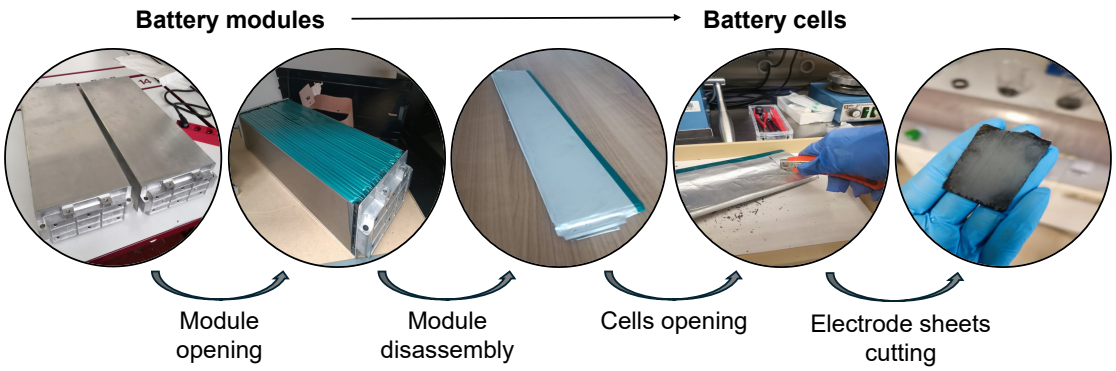

Figure 2.1: Illustration of lithium-ion battery module disassembly. Further details are provided in works [C5] and [J2].

The findings highlight the complexities involved in the manual disassembly module process, contributing to a refined understanding of the steps necessary for process optimisation and automation, which is crucial for improving the efficiency of subsequent material recovery in LIB recycling.

Recognising the limitations of conventional methods in achieving efficient and environmentally friendly separation, the ultrasonic-assisted technique has been explored in [C6] as a promising pre-treatment option for sustainable material recovery. This study demonstrates the effectiveness of ultrasonic separation for detaching electrode materials from NMC 622 pouch cells using various solvents, including ethanol, acetone, isopropyl alcohol, N-Methyl-2-pyrrolidone (NMP), and distilled water, achieving separation efficiencies as shown in Fig. 2.2. Among these, distilled water yielded high detachment rates (69.4% for the positive electrode, 72.6% for the negative electrode) while offering significant environmental benefits. As a non-toxic and non-flammable solvent, it reduces chemical waste and eliminates the need for energy-intensive solvent recovery processes.

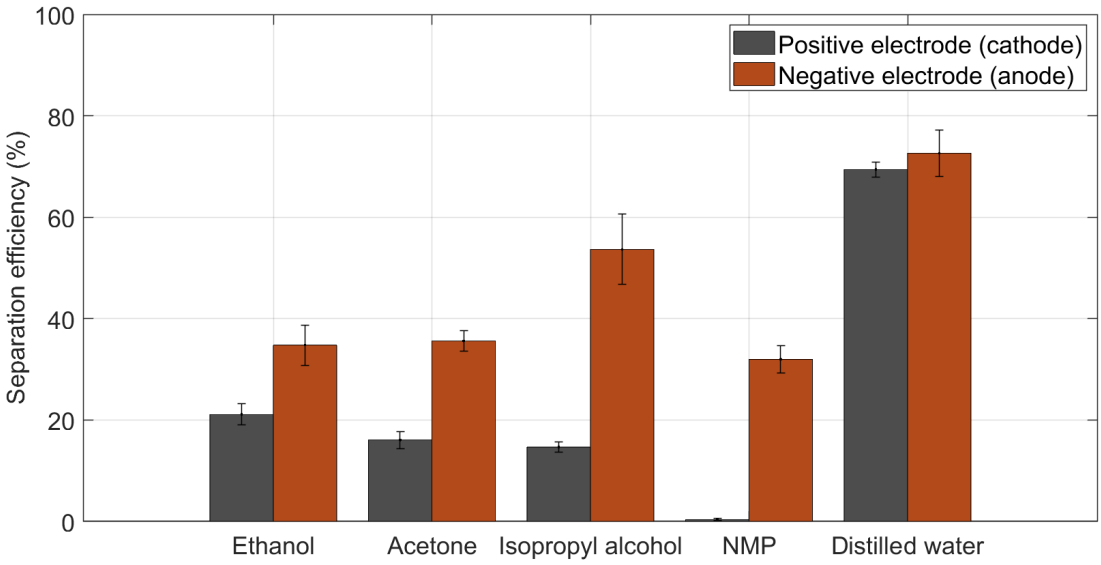

Figure 2.2: A comparison of the process efficiencies in ultrasonic-assisted material separation of lithium-ion battery pouch cell electrodes using different solvents. NMP = N-Methyl-2-pyrrolidone. The terms 'cathode' and 'anode' refer to the positive and negative electrodes following the battery's discharging process [4]. Further details are provided in the work [C6].

However, the study also reveals challenges in scaling up the process for industrial applications, specifically a reduction in separation efficiency with increased sample loads, which calls for optimised agitation and solvent delivery systems. The findings underscore the potential of ultrasonic separation to contribute to more environmentally friendly and efficient LIB recycling, provided that further research and engineering solutions address the scalability and throughput limitations for industrial adoption.

Furthermore, a detailed material characterisation of deeply discharged pouch LIB cells (3.65 V, 78 Ah) from a BEV was presented in [J2]. The work detailed the material composition of the NMC 622 battery cell, including a $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ cathode

on an aluminium current collector, a graphite anode on a copper foil, a polyethylene (PE)/ polyvinylidene fluoride (PVDF) polymer separator, and a lithium hexafluorophosphate ($LiPF_6$) salt electrolyte with a 1:3 ratio in primary solvents DMC and DEC. The characterisation involved a series of X-ray and infrared spectroscopy (IRS) measurements, and GC-MS coupled with ICP-OES to analyse the electrolyte solution. Such detailed material analyses are indispensable for optimising downstream recycling processes, enabling accurate quantification of valuable material content and contaminants, and facilitating the design of tailored pre-treatment and separation methodologies to yield higher quality BM.

Complementing these studies, the thermal stability of valuable metals in EOL LIB cathode material ($LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$) was investigated in [J3]. This study, which is focused on a temperature range of 100–400 °C, which corresponds to the standard thermal pre-treatment conditions employed in recycling processes [30], [86], [87], utilised ICP-OES to quantify the concentrations of lithium, nickel, manganese, and cobalt leached from the cathode material after annealing at the specified temperatures. XRF, XRD, thermogravimetric analysis (TGA), and SEM with EDS were employed to assess the material's properties and structural changes following thermal treatment. The key findings indicate that a temperature of 400 °C yields the highest leaching efficiency of valuable metals when combined with dissolution in 20% nitric acid for 4 hours. These results contribute to refining pre-treatment parameters in LIB recycling, with implications for improving the quality and recovery of valuable metals while reducing energy consumption and minimising environmental impact.

Collectively, presented works contribute to a broadening knowledge base focused on enhancing LIB recycling efficiency through improved BM quality and metal recovery. Integrating advanced characterisation techniques, optimised disassembly strategies, and innovative separation approaches, including ultrasonic-assisted detachment and thermal pre-treatment, represents a concerted effort to address key challenges in the field.

To contextualise these efforts further and highlight the importance of enhancing pre-treatment processes, the review work [J4] provides a comprehensive overview of the limitations of current direct recycling methods, which are directly linked to BM processing, as described in Tab. 2.1. As emphasised, a key challenge lies in the effective processing of BM derived from LIB recycling pre-treatment, which often contains impurities that hinder direct recycling efficiency. This review underscores that optimising pre-treatment to obtain high-quality BM or electrode pre-products is crucial for successfully implementing direct recycling technologies. Notably, the review emphasises that inadequate pre-treatment steps lead to increased energy consumption and environmental burdens due to additional treatments required to remove impurities.

However, continued research is imperative to refine process parameters further, establish the scalability of sustainable separation methods, and develop automation strategies that underpin a robust and economically viable LIB recycling industry within a circular battery economy framework. Future work should mainly focus on the interplay between pre-treatment methods and their effects on BM's chemical and physical properties, to enable the development of tailored downstream processes that maximise material recovery and minimise waste.

Table 2.1: Overview of main limitations of currently implemented direct recycling techniques. Reprinted from [J4] under the terms and conditions of the CC BY 4.0. © 2024.

Limitation	Description
Limited applicability to whole spent LIB systems	The current approaches predominantly target the cathode and anode materials, neglecting other components of spent LIBs; thus, the potential for achieving a closed-loop circular economy for LIBs is not fully harnessed.
Black mass processing	The standard black mass, comprising a mixture of several materials, introduces technical barriers to the direct implementation of direct recycling techniques.
Energy-intensive recycling pretreatment	Before direct recycling, LIBs undergo recycling pretreatment processes involving high-temperature treatments and chemical interventions, which result in high energy consumption, emissions, and intricacies in the process of wastewater treatment.

2.2 Conclusions and Perspectives

The presented work highlights the crucial role of pre-treatment in LIB recycling, particularly its influence on BM quality and overall process efficiency. Exploring and assessing advanced strategies for disassembly, material characterisation, and separation reflects a broader shift toward more refined and sustainable recycling approaches. Laboratory-scale analytical methods, including SEM-EDS, XRF, XRD, and ICP-OES, were employed to investigate battery components' chemical and structural composition. These techniques yielded valuable insights into material behaviour during pre-treatment, enabling a more comprehensive evaluation of process effectiveness. While not intended for direct industrial application, the outcomes contribute to developing better-informed strategies for improving BM purity and maximising resource recovery.

Despite current progress, critical challenges remain in the recycling pre-treatment field. Many promising techniques, such as ultrasonic-assisted separation or solvent-based treatments, still face scalability, process consistency, or economic feasibility limitations. Manual disassembly and cell handling also introduce safety risks and labour constraints, particularly in high-throughput industrial settings. Automation and robotics offer a way forward, but must be carefully tailored for battery design variability and diverse chemistries.

Future research should aim to refine pre-treatment methodologies to reduce BM impurity levels, support direct recycling development, and enhance overall material recovery. Special attention should be paid to the interplay between pre-treatment steps and the resulting chemical and physical properties of processed materials. Addressing these interactions will be essential to designing recycling strategies that are both efficient and adaptable to evolving battery technologies.

Chapter 3

Comprehensive Evaluation of Lithium-Ion Battery Recycling Pre-Treatment Processes

The EOL management of LIBs presents economic opportunities and environmental challenges, with the pre-treatment phase playing a pivotal role in determining the efficiency and sustainability of material recovery [8], [88], [89]. Addressing its challenges requires technical advancements [90] and comprehensive environmental and economic assessments to optimise recovery rates, reduce processing costs, and maximise overall profitability while minimising ecological impact [28], [91], [92]. Beyond improving material yield and purity, effective pre-treatment strategies can reduce waste generation, lower emissions, and enhance resource efficiency, contributing to a more sustainable recycling process [29]. Moreover, optimising these processes reduces operational costs, ultimately shaping the overall lifecycle economics of LIB systems [47].

Each step of pre-treatment carries distinct economic and environmental implications [29]. Battery sorting enhances the quality of the BM and recovered materials by enabling more targeted processing [49], [93]. However, industrial-scale sorting systems capable of accurately classifying all LIB types remain costly, and their slow processing speeds further hinder widespread implementation [49].

Before further processing, the discharging of LIBs represents a critical safety measure to prevent thermal runaway and short-circuiting during handling [42], [90], [94]. Electrochemical discharging methods generate contaminated wastewater and potentially harmful gas emissions, necessitating additional treatment to mitigate ecological damage [52], [74]. Their economic viability depends on the cost of chemical reagents and energy requirements for solution preparation and waste treatment [76]. In contrast, non-electrochemical discharging techniques eliminate chemical waste but may pose safety concerns due to heat generation, requiring controlled environments and increasing infrastructure costs [30], [76]. While specialised systems can enable energy recovery during discharging, their adoption is often limited by high initial investments and technical constraints, making widespread implementation challenging [29], [76].

Regarding dismantling, the manual approach allows for a high degree of separation but is labour-intensive and exposes workers to hazardous materials [51]. In contrast, automated dismantling enhances efficiency and safety but requires substantial capital investment [80]. Both approaches necessitate strict ventilation and safety protocols, adding to operational costs [50], [51], [80]. Comminution, in which batteries are crushed to liberate electrode materials, is energy-intensive and generates particulate matter, requiring dust control measures to mitigate respiratory hazards [57]. The associated energy demands and emissions contribute to economic and environmental costs [29], [56], [58].

Classification, typically achieved through sieving, is an energy-intensive process that generates dust, requiring filtration systems to minimise environmental impact [63], [95]. This step incurs costs primarily related to the operation and maintenance of the equipment. Similarly, separation processes isolate valuable materials, introduce significant energy, and generate secondary waste [95], [96]. For instance, while froth flotation is efficient, it uses chemical reagents that must be handled carefully to prevent environmental contamination [97]. The economic viability of these separation methods depends on factors such as equipment complexity, reagent costs, and overall energy demand, which significantly affect the cost-effectiveness of the recycling process [29], [96].

Chemical and thermal treatments play a significant role in material recovery but also introduce several challenges [84], [98]. Chemical methods, such as solvent dissolution, efficiently remove binders but raise environmental concerns due to solvent toxicity and volatility, necessitating stringent safety measures and recovery systems [98], [99]. Similarly, thermal treatments like pyrolysis are energy-intensive and generate emissions, requiring effective emission control technologies [84]. The costs associated with these and previously discussed processes depend on multiple factors, including overall energy consumption, chemical reagent use, environmental compliance requirements, waste treatment, equipment maintenance, labour expenses, and the need for specialised infrastructure [29].

Improving pre-treatment processes will enhance economic viability and environmental performance as LIB recycling evolves. Innovations in automation, process efficiency, and the integration of cleaner energy can help address existing limitations, reducing costs and minimising ecological impacts. Future developments should concentrate on adaptive and scalable solutions that can accommodate the growing volume and diversity of EOL batteries, ensuring that recycling remains sustainable and viable, both technologically and economically, in the long term.

3.1 Author's Contribution to This Research Field

In summary, each pre-treatment step in LIB recycling presents a trade-off between economic feasibility and environmental responsibility. Optimising these steps to minimise costs and environmental impacts is crucial for establishing a sustainable and economically viable LIB recycling industry. While various studies [3], [100]–[102] assess the efficiency of different pre-treatment methods, it is essential to recognise that such evaluations are often conducted under specific operational conditions, influenced by factors such as facil-

ity infrastructure, technological expertise, waste management strategies, and regulatory frameworks. Consequently, no single pre-treatment approach can be universally deemed superior. The overall effectiveness of a given method depends on its implementation within a particular system, including its integration with downstream processing and waste handling. Therefore, an evaluation tailored to the specific operational context is essential to accurately assess any pre-treatment process's economic and environmental viability.

A study in [J5] consolidates existing techno-environmental and economic research of LIB recycling and provides a comprehensive foundation for academic studies and industrial applications. The review identifies key challenges and opportunities in the field by synthesising current knowledge on pre-treatment and recycling methods. It underscores the need for further research to enhance process efficiency, reduce environmental burdens, and improve the economic value of recovered materials. A particular emphasis is placed on the role of LCA and other environmental indicators in evaluating the sustainability of LIB recycling. The review highlights how recycling can significantly conserve valuable resources, reduce dependence on primary extraction, and lower greenhouse gas emissions. The comparative assessment of pyrometallurgy, hydrometallurgy, and direct recycling demonstrates the varying environmental footprints of these techniques, informing the selection of optimal recycling strategies.

The economic perspective of LIB recycling is explored through works presenting a cost-benefit analysis, considering investment and operational costs alongside the revenue potential of recovered materials. The review emphasises the circular economy framework to maximise resource efficiency and minimise waste. Factors influencing profitability, such as battery chemistry, material prices, technological advancements, and regional economic conditions, are examined to determine the feasibility of different recycling approaches. Ultimately, the study concludes that effective LIB recycling is essential for the sustainable development of the EV industry, necessitating an integrated approach that balances environmental responsibility with economic viability.

Despite the limited research focus on EOL management and recycling of LIBs in the Czech Republic, this study [C7] serves as an initial step in addressing this gap. It provides a foundational assessment by estimating the recycling flows of waste LIBs from EVs in the Czech Republic between 2020 and 2025. This estimation is crucial for subsequent research on optimising recycling strategies and infrastructure. The study develops a predictive model to quantify material recovery from two LIB chemistries—lithium manganese oxide (LMO) and NMC—across three EV categories: BEVs, plug-in hybrid electric vehicles (PHEVs), and hybrid electric vehicles (HEVs). Furthermore, it evaluates different recycling rate scenarios based on the current legislative framework. It offers insights into optimising recycling facilities and strategies for reducing dependence on primary raw materials by reusing recovered materials.

The findings highlight key trends in electromobility within the Czech Republic, which are essential for designing recycling infrastructure and waste-material logistics. The study projected that between 2020 and 2025, the number of BEVs would be approximately 2.5 times higher than that of PHEVs, with the total EV fleet reaching 40,000 by 2025. However, a comparison with current statistics reveals a deviation from these initial pro-

jections. Actual vehicle registration data from the end of 2024 [103] show that the total EV fleet (BEV + PHEV) surpassed the forecast, reaching approximately 56,000 vehicles. Furthermore, the actual ratio of BEVs (approx. 32,000) to PHEVs (approx. 24,000) stood at roughly 1.33, considerably lower than the projected 2.5, indicating stronger-than-anticipated growth in the PHEV segment alongside BEVs. The increasing number of EVs, which is growing even faster than initially projected, will inevitably lead to a rise in EOL LIB waste. Legislative measures tightening recycling requirements, including an increase in the recycling rate from 50% to 65% by the end of 2025 [25], are anticipated to enhance materials recovery significantly. While the specific quantitative forecasts differed from the observed outcomes, the initial analysis correctly identified the significant upward trend in EV adoption and its crucial implications for EOL battery management.

In the study [J6], the environmental assessment of LIB recycling pre-treatment in the conditions of the Czech Republic was conducted through a partial LCA study, evaluating the environmental impacts associated with processing three distinct battery chemistries: LCO/NMC, NMC, and LFP. The work primarily focuses on the production of BM as the main output and highlights the substantial recovery of secondary aluminium as a key environmental benefit. Notably, the findings indicated that the varying chemistry of these LIBs had no significant effect on the overall environmental impacts of the pre-treatment process. The study emphasised that the prevention of primary aluminium production, which is highly energy- and resource-intensive, constitutes a significant advantage of LIB recycling. These results provided a foundational understanding of the environmental implications of LIB recycling pre-treatment. They underscored the need for further research to optimise process efficiency and expand the scope of impact assessments.

The following work [J7] builds upon a previous study by extending the scope of LIB recycling pre-treatment in the Czech Republic to include both CECs and EV batteries. A key advancement lies in evaluating different discharging methods—electrochemical for CECs and active discharging for EV batteries, ensuring a tailored approach to waste processing (Fig. 3.1). This study employs a gate-to-gate LCA using SimaPro and Ecoinvent 3.9 to assess environmental impacts across multiple categories, including climate change, eutrophication, freshwater use, and resource depletion. The findings highlight the crucial role of secondary material recovery, particularly for copper and aluminium, in reducing resource consumption and emissions.

Results indicate that CEC recycling demonstrates slightly higher climate change mitigation benefits than EV battery recycling. However, the pre-treatment process for EV batteries contributes more significantly to resource conservation, achieving a 55% reduction in mineral and metal use compared to production impacts. The study underscores the importance of optimising pre-treatment processes to enhance recycling efficiency and minimise environmental burdens. Furthermore, it emphasises the role of a cleaner energy mix in reducing overall impacts, reinforcing the need for policy-driven transitions in battery recycling infrastructure. These insights provide a foundation for future research into advanced recycling technologies and their integration into sustainable energy systems.

The study in [J8] highlights the importance of addressing EOL treatment strategies for BESSs. Focused on a 2.8 MWh/2.5 MW BESS, the research evaluates the LIB system's

recycling pre-treatment, cooling and extinguishing systems, inverters, and the reuse of containers and substations in new BESS installations. Environmental benefits were assessed using a gate-to-gate LCA, measuring impacts with the most critical categories, such as climate change, freshwater eutrophication, and resource use. Illustration of LCA results for the LIB system treatment is provided in Fig. 3.2. The findings reveal significant environmental advantages, primarily due to the recovery of secondary materials like aluminium and copper, which account for approximately 50% of the environmental benefits. The recovery of secondary aluminium from batteries, cooling systems, and inverters is a key factor in reducing the carbon footprint of BESS recycling, while recycling fire extinguishers yields mainly secondary steel.

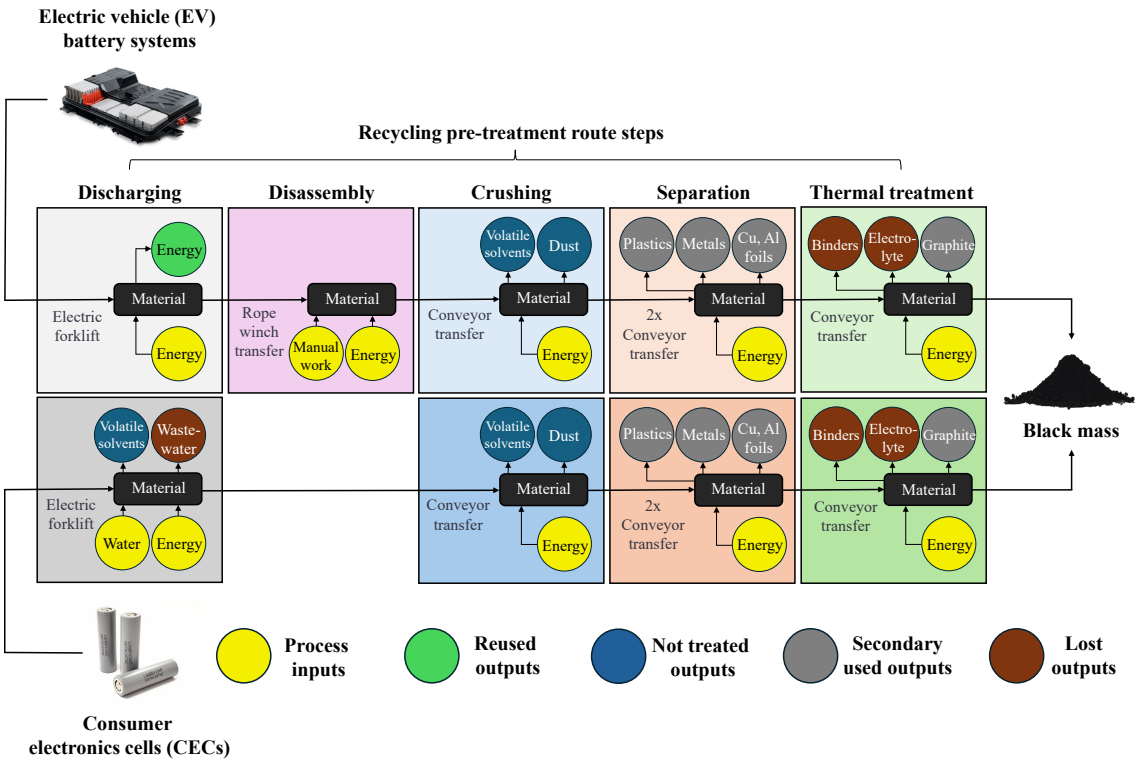

Figure 3.1: Process scheme of the recycling pre-treatment steps for spent lithium-ion batteries from electric vehicles (EVs) and consumer electronics. Further details are provided in the work [J7].

Economically, the study identifies nickel as the most significant contributor to profits, followed by cobalt, aluminium, and copper. Together, these materials account for over 90% of the projected profits from recycling. The study also emphasises the potential value of BESS containers at EOL, which can mitigate investment risks and support a transition to greener energy. Based in the Czech Republic, the methodology offers global applicability, demonstrating the environmental and economic benefits of recycling, local sourcing, and

reduced import reliance. Transitioning to renewable energy sources for recycling would further optimise these benefits, aligning with sustainability goals and promoting an eco-friendlier approach to BESS EOL treatment.

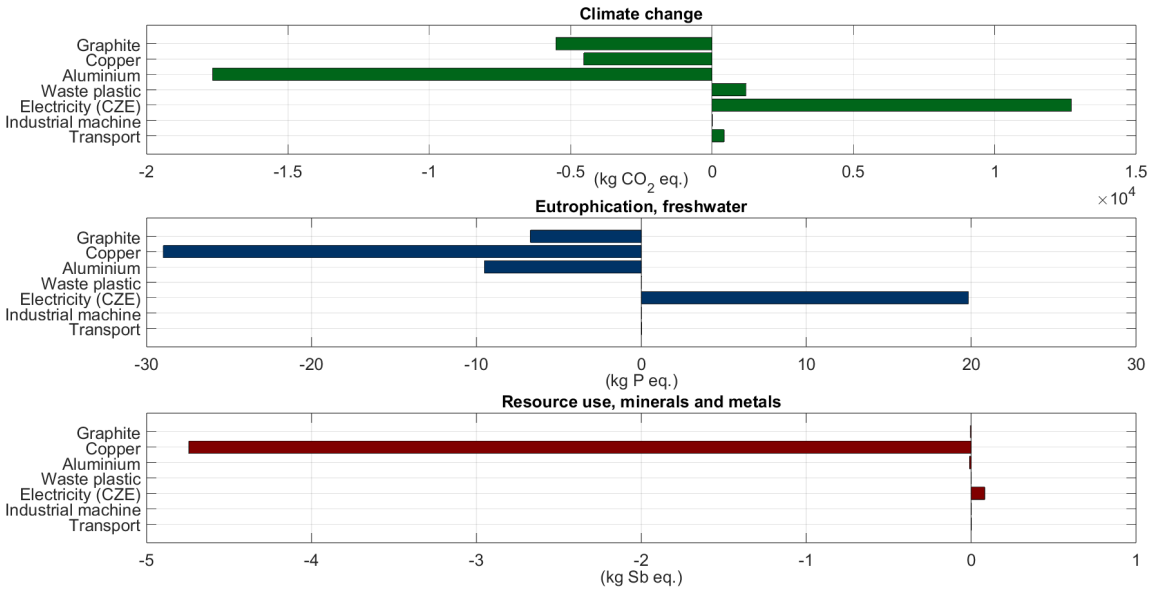

Figure 3.2: Environmental impact categories assessed through a partial life cycle assessment (LCA) for the end-of-life treatment of a lithium-ion battery system used in a battery energy storage system (BESS) installation. Results include impacts related to climate change, eutrophication, freshwater use, and resource use (minerals and metals). Further details are provided in the work [J8].

As a further complement to this research field, a study [C8] evaluates the economic feasibility of LIB recycling from EVs in the Czech Republic, focusing on a combined pyrometallurgical and hydrometallurgical process. The study examines the costs associated with different recycling steps, considering factors such as process efficiency (above 95%) and the high quality of recovered materials. Two EV battery capacities—32 kWh (small vehicle) and 80 kWh (large vehicle)—are analysed to estimate recycling expenses. Additionally, the study explores future scenarios, projecting the economic impact of anticipated EU legislative changes over the next 5 and 10 years.

The results indicate that establishing local recycling plants could significantly reduce the current cost of LIB recycling in the Czech Republic, which relies on transport to recycling facilities in Germany. Future projections suggest that a Polish facility could lower transportation costs within five years, while a domestic Czech facility within ten years could further optimise expenses. The study also considers technological advancements that could increase process efficiency and offset rising energy costs. These insights can inform investment decisions, such as return-on-investment calculations for new recycling facilities and contribute to developing sustainable and economically viable LIB recycling strategies.

3.2 Conclusions and Perspectives

This work provides a comprehensive overview of LIB recycling pre-treatment's environmental and economic implications, emphasising this evolving sector's challenges and significant potential. The findings highlight the dual benefits of pre-treatment: the reduction of environmental burdens and creating economic value. The research illustrates the necessity of tailoring recycling strategies to specific operational, regional, and regulatory contexts by combining partial LCAs and techno-economic evaluations.

While different strategies present trade-offs between economic feasibility and environmental impact, the results underscore the importance of adapting pre-treatment processes to local infrastructure, energy mix, and material composition. The integration of LCA and techno-economic assessments has provided valuable insights into the sustainability of various approaches, reinforcing the role of secondary material recovery, particularly aluminium, copper, and nickel, in reducing resource depletion and emissions.

Moreover, the presented studies reflect the evolving landscape of LIB recycling in the Czech Republic, where infrastructure development and policy changes will be key to shaping the industry's economic and environmental outlook. The assessment of pre-treatment strategies contributes to a broader understanding of EOL battery management. The economic feasibility evaluation also highlights the urgent need for localised recycling facilities to improve cost efficiency and reduce dependence on cross-border processing.

Future research should focus on enhancing the efficiency and selectivity of pre-treatment technologies to maximise material recovery, minimise secondary waste, and reduce overall energy demand. Further integration of cleaner energy sources into recycling operations and advancements in hybrid or alternative pre-treatment technologies could significantly enhance the sector's sustainability. Establishing regional infrastructure and closing material loops locally will be crucial in making LIB recycling environmentally sound and economically viable. Addressing these key aspects will help LIB recycling transition toward a circular economy model, ensuring environmental responsibility and long-term economic resilience.

Conclusion

This dissertation highlights the critical role of pre-treatment processes in LIB recycling, emphasising their impact on efficiency, sustainability, and economic viability. The research successfully addressed three key areas: electrochemical discharging, BM quality optimisation to enhance material recovery, and the systematic evaluation of pre-treatment methodologies to balance economic and environmental factors.

The study of electrochemical discharging confirms its critical role in reducing residual voltage in EOL LIBs while revealing considerable environmental and economic challenges. This procedure often releases hazardous compounds and metal contamination into wastewater, requiring comprehensive treatment strategies. Additionally, variability in discharging efficiency may lead to material losses and increased operational costs, underlining the urgent need for process optimisation and improved waste handling. The newly proposed non-invasive discharging method has emerged as a promising alternative in response to these limitations. It provides comparable discharge durations and efficiencies, producing fully discharged cells safe for further processing, while providing 100% elimination of electrolyte leakage and completely preventing the release of key components such as DMC, EMC, and DEC into the coolant. This complete absence of electrolyte leakage, in contrast to conventional electrochemical methods, directly demonstrates the preservation of cell integrity by preventing mechanical damage to the casing. These findings directly support the first research objective and clarify the potential, limitations, and practical demands of aqueous discharging strategies.

Regarding BM quality optimisation, the findings demonstrate the profound impact of pre-treatment on material composition and purity. Inefficiencies in sorting, dismantling, comminution, and separation contribute to contamination, resource losses, and heightened energy demands, ultimately hindering downstream recovery. This dissertation explores and experimentally validates novel separation approaches, including ultrasonic-assisted separation resulting in detachment rates of up to 69.4% for the positive electrode and 72.6% for the negative electrode using distilled water, enhancing recovery rates and minimising environmental impact by avoiding harsh solvents. Moreover, the thermal conditioning protocol demonstrated that 400 °C is sufficient for effective metal leaching, further contributing to improved resource recovery and reduced energy consumption. Such developments, proposed and tested as part of this work, significantly contribute to current pre-treatment practices. By proposing more sustainable and effective alternatives, the work addresses the second research objective and provides practical pathways for improving the quality and consistency of intermediate battery materials.

The evaluation of pre-treatment strategies, specifically for the Czech Republic, using LCA and techno-economic assessments, highlights the trade-off between economic feasibility and environmental responsibility. The results show that CEC recycling offers slightly greater benefits for climate-change mitigation than EV battery recycling. However, the pre-treatment process for EV batteries contributes more significantly to resource conservation, achieving a 55% reduction in mineral and metal use compared to production impacts. These findings underscore the environmental advantages of LIB recycling, especially in cutting emissions and conserving resources. For BESSs, secondary-material recovery—particularly of aluminium and copper—accounts for approximately 50% of the environmental benefits. In contrast, nickel, cobalt, aluminium, and copper contribute over 90% of projected recycling profits. Economic analyses confirm the profitability of these recovery processes, reinforcing the need for region-specific infrastructures to enhance cost-effectiveness and sustainability. This integrative approach meets the third research objective and contributes to regional policymaking and infrastructure planning.

This work provides a comprehensive, experimentally grounded, and sustainability-oriented perspective on LIB pre-treatment, underscoring the necessity of integrating efficient and environmentally responsible processes to advance LIB recycling towards a circular economy. The research objectives defined at the outset have been fulfilled through targeted investigations and interdisciplinary analyses, resulting in several original contributions to scientific understanding and applied methodologies, including:

- Demonstration of key environmental and economic challenges associated with conventional electrochemical battery discharging in aqueous media.
- Development and testing of a novel non-invasive discharging method that enhances safety and prevents electrolyte leakage.
- Identification of critical inefficiencies in sorting, dismantling, comminution, and separation during pre-treatment.
- Exploration and experimental validation of ultrasonic-assisted separation as a more sustainable alternative to solvent-based processing.
- Integration of life-cycle and techno-economic assessments demonstrates the trade-offs between environmental benefits and economic feasibility and highlights the need for tailored regional recycling infrastructure.

These contributions collectively advance the understanding of LIB pre-treatment and provide a foundation for developing more sustainable and economically viable recycling.

Future efforts should focus on scaling up automated and sustainable pre-treatment technologies, optimising waste management frameworks, and fostering policy-driven incentives to support industry-wide adoption. These conclusions serve as academic insight and practical guidance for further development of LIB recycling systems. By addressing these challenges, LIB recycling can evolve into a more resilient, economically viable, and environmentally sustainable process.

Author's Bibliography

The list of publications and patents presents, for each entry, the percentage contributions of all authors and inventors, citation counts from Web of Science (WoS), Scopus, and Google Scholar (GS), and the most recent impact factors (IFs) of the respective journals at the time of submission. Citation data were collected on April 26, 2025.

Core Publications and Patents

Publications in Peer-reviewed Journals with Impact Factor

[J1] **A. Pražanová**, V. Knap, and D.-I. Stroe, "Literature Review, Recycling of Lithium-Ion Batteries from Electric Vehicles, Part I: Recycling Technology," *Energies 2022, Vol. 15, Page 1086*, vol. 15, p. 1086, 3 Feb. 2022, ISSN: 1996-1073. DOI: 10.3390/EN15031086, **Contributions: AP: 33.33%, VK: 33.33%, DIS: 33.33%, IF 3.0 (Q3 in Energy and Fuels), citations: 34 in WoS, 45 in Scopus, 65 in GS.**

[J2] **A. Pražanová**, J. Kočí, M. H. Míka, D. Pilnaj, Z. Plachý, and V. Knap, "Pre-Recycling Material Analysis of NMC Lithium-Ion Battery Cells from Electric Vehicles," *Crystals 2023, Vol. 13, Page 214*, vol. 13, p. 214, 2 Jan. 2023, ISSN: 2073-4352. DOI: 10.3390/CRYST13020214, **Contributions: AP: 16.67%, JK: 16.67%, MHM: 16.67%, DP: 16.67%, ZP: 16.67%, VK: 16.67%, IF 2.4 (Q2 in Crystallography), citations: 7 in WoS, 7 in Scopus, 9 in GS.**

[J3] N. Klusoňová, E. Sedláčková, J. Kočí, D. Pilnaj, K. Pánová, J. Uřičář, V. Procházka, K. Jílková, **A. Pražanová**, and M. H. Míka, "Thermal Stability of Valuable Metals in Lithium-Ion Battery Cathode Materials: Temperature Range 100–400 °C," *Journal of Power Sources*, vol. 627, p. 235 795, Jan. 2025, ISSN: 0378-7753. DOI: 10.1016/J.JPOWSOUR.2024.235795, **Contributions: NK: 10%, ES: 10%, JK: 10%, DP: 10%, KP: 10%, JU: 10%, VP: 10%, KJ: 10%, AP: 10%, MHM: 10%, IF 8.1 (Q1 in Electrochemistry), citations: 2 in WoS, 2 in Scopus, 2 in GS.**

[J4] **A. Pražanová**, Z. Plachý, J. Kočí, M. Fridrich, and V. Knap, "Direct Recycling Technology for Spent Lithium-Ion Batteries: Limitations of Current Implementation," *Batteries 2024, Vol. 10, Page 81*, vol. 10, p. 81, 3 Feb. 2024, ISSN: 2313-0105. DOI: 10.3390/BATTERIES10030081, **Contributions: AP: 20%, ZP: 20%, JK: 20%, MF: 20%, VK: 20%, IF 4.6 (Q2 in Electrochemistry), citations: 7 in WoS, 10 in Scopus, 13 in GS.**

[J5] **A. Pražanová**, V. Knap, and D.-I. Stroe, "Literature Review, Recycling of Lithium-Ion Batteries from Electric Vehicles, Part II: Environmental and economic perspective," *Energies 2022, Vol. 15, Page 7356*, vol. 15, p. 7356, 19 Oct. 2022, ISSN: 1996-1073. DOI: 10.3390/EN15197356, **Contributions: AP: 33.33%, VK: 33.33%, DIS: 33.33%, IF 3.0 (Q3 in Energy and Fuels), citations: 29 in WoS, 31 in Scopus, 38 in GS.**

[J6] M. Fridrich, **A. Pražanová**, J. Weinzettel, and V. Knap, "Lithium-Ion (LCO/NMC, NMC, LFP) Battery Recycling: Partial LCA Study," *Monatshefte fur Chemie*, vol. 155, pp. 309–312, 3-4 Apr. 2024, ISSN: 00269247. DOI: 10.1007/S00706-024-03184-6/TABLES/2, **Contributions: MF: 25%, AP: 25%, JW: 25%, VK: 25%, IF 1.7 (Q3 in Chemistry, Multidisciplinary), citations: 1 in WoS, 1 in Scopus, 1 in GS.**

[J7] **A. Pražanová**, M. Fridrich, J. Weinzettel, and V. Knap, "Gate-to-Gate Life Cycle Assessment of Lithium-Ion Battery Recycling Pre-Treatment," *Cleaner Environmental Systems*, vol. 16, p. 100 263, Mar. 2025, ISSN: 2666-7894. DOI: 10.1016/J.CESYS.2025.100263, **Contributions: AP: 25%, MF: 25%, JW: 25%, VK: 25%, IF 6.1 (Q1 in Environmental Science), citations: 0 in WoS, 0 in Scopus, 0 in GS.**

[J8] **A. Pražanová**, Z. Plachý, M. Fridrich, *et al.*, "Environmental and Economic Assessment of Battery Energy Storage System Disposal and Recycling Treatment in the Czech Republic," submitted for review, Feb. 2025., **Contributions: AP: 14.29%, ZP: 14.29%, MF: 14.29%, DH: 14.29%, JW: 14.29%, DIS: 14.29%, VK: 14.29%.**

Patents

[P1] **A. Pražanová**, Z. Plachý, and V. Knap, *Způsob vybíjení bateriového článku*, Czech patent, CZ2024247A3, 2024, Also filed as European patent application EP25157404.2 on 12 Feb. 2025, **Contributions: AP: 46%, ZP: 46%, VK: 8%.**

Conference Contributions

[C1] **A. Pražanová**, D. Pilnaj, Z. Plachý, and V. Knap, "Techno-Environmental Evaluation of Recycling Pretreatment of Cylindrical Lithium-Ion Battery: Discharging via Salt-Based Solution," *46th International Spring Seminar on Electronics Technology (ISSE 2023)*, pp. 1–4, May 2023. DOI: 10.1109/ISSE57496.2023.10168348, **Contributions: AP: 25%, DP: 25%, ZP: 25%, VK: 25%, citations: N/A in WoS, 3 in Scopus, 3 in GS.**

[C2] **A. Pražanová**, Z. Plachý, J. Uřičář, D. Pilnaj, J. Kočí, and V. Knap, "Sustainability Challenges: The Circular Economy Dilemma in Lithium-Ion Battery Cell Electrochemical Discharging Processes," *Proceedings of the International Spring Seminar on Electronics Technology (ISSE 2024)*, 2024, ISSN: 21612536. DOI: 10.1109/ISSE61612.2024.10603724, **Contributions: AP: 16.67%, ZP: 16.67%, JU: 16.67%, DP: 16.67%, JK: 16.67%, VK: 16.67%, citations: 0 in WoS, 1 in Scopus, 2 in GS.**

[C3] **A. Pražanová**, D. Pilnaj, M. H. Míka, and V. Knap, "Lithium-Ion NMC Batteries: Chemical Toxicity Reflection of Wastewater and Scrap," in *The 6th International Conference on Nanomaterials: Fundamentals and Applications*, Slovenská chemická společnosť při SAV, 2022, pp. 118–121, **Contributions: AP: 25%, DP: 25%, MHM: 25%, VK: 25%, citations: N/A in WoS, N/A in Scopus, N/A in GS.**

[C4] J. Kočí, **A. Pražanová**, Z. Plachý, and V. Knap, "Unlocking Insights: A Systematic Survey of Material Composition in Lithium-Ion Battery Cells for Recycling Solutions," *Proceedings of the International Spring Seminar on Electronics Technology*, 2024, ISSN: 21612536. DOI: 10.1109/ISSE61612.2024.10603575, **Contributions: JK: 25%, AP: 25%, ZP: 25%, VK: 25%, citations: 0 in WoS, 0 in Scopus, 0 in GS.**

[C5] **A. Pražanová**, M. H. Míka, and V. Knap, "Lithium-Ion Battery Module-to-Cell: Disassembly and Material Analysis," *Journal of Physics: Conference Series*, vol. 2382, p. 12 002, 2022. DOI: 10.1088/1742-6596/2382/1/012002, **Contributions: AP: 33.33%, MHM: 33.33%, VK: 33.33%, citations: N/A in WoS, 5 in Scopus, 5 in GS.**

[C6] **A. Pražanová**, Z. Plachý, J. Kočí, D. Havlíček, and V. Knap, "Ultrasonic-Assisted Electrode Material Separation: Towards Industrial Applications," in *48th International Spring Seminar on Electronics Technology (ISSE 2025)*, accepted from April 2025, **Contributions: AP: 20%, ZP: 20%, JK: 20%, DH: 20%, VK: 20%.**

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[C8] **A. Pražanová** and V. Knap, "Ekonomická náročnost recyklace lithium-iontových baterií z elektromobilů," in *43. Nekonvenční zdroje elektrické energie*, Česká elektrotechnická společnost, 2022, pp. 95–98, **Contributions: AP: 50%, VK: 50%, citations: N/A in WoS, N/A in Scopus, N/A in GS.**

Author's Other Publications

Publications in Peer-reviewed Journals

[OJ1] K. Dušek, D. Bušek, P. Veselý, **A. Pražanová**, M. Plaček, and J. D. Re, "Understanding the Effect of Reflow Profile on the Metallurgical Properties of of Tin–Bismuth Solders," *Metals 2022, Vol. 12, Page 121*, vol. 12, p. 121, Jan. 2022, ISSN: 2075-4701. DOI: 10.3390/MET12010121, **Contributions: KD: 25%, DB: 25%, PV: 25%, AP: 10%, MP: 10%, JDR: 5%, IF 2.6 (Q2 in Metallurgy & Metallurgical Engineering), citations: 12 in WoS, 14 in Scopus, 17 in GS.**

[OJ2] K. Nováková, **A. Pražanová**, D.-I. Stroe, and V. Knap, "Second-Life of Lithium-Ion Batteries from Electric Vehicles: Concept, Aging, Testing, and Applications," *Energies 2023, Vol. 16, Page 2345*, vol. 16, p. 2345, 5 Feb. 2023, ISSN: 1996-1073. DOI: 10.3390/EN16052345, **Contributions: KN: 25%, AP: 25%, DIS: 25%, VK: 25%, IF 3.0 (Q3 in Energy and Fuels), citations: 17 in WoS, 18 in Scopus, 23 in GS.**

[OJ3] L. Gdula, B. Dzurňák, **A. Pražanová**, and T. Markvart, "ULTRATHIN ORGANIC DYE LAYERS FOR SENSITISATION OF SILICON," *Journal of Chemical Technology and Metallurgy*, vol. 59, pp. 329–334, 2 Jan. 2024, ISSN: 1314-7978. DOI: 10.59957/JCTM.V59.I2.2024.11, **Contributions: LG: 40%, BD: 30%, AP: 20%, TM: 10%, citations: N/A in WoS, 0 in Scopus N/A in GS.**

[OJ4] E. Sedláčková, **A. Pražanová**, Z. Plachý, N. Klusoňová, V. Knap, and K. Dušek, "Acoustic Emission Technique for Battery Health Monitoring: Comprehensive Literature Review," *Batteries 2025, Vol. 11, Page 14*, vol. 11, p. 14, 1 Jan. 2025, ISSN: 2313-0105. DOI: 10.3390/BATTERIES11010014, **Contributions: ES: 16.67%, AP: 16.67%, ZP: 16.67%, NK: 16.67%, VK: 16.67%, KD: 16.67%, IF 4.6 (Q2 in Electrochemistry), citations: 0 in WoS, 0 in Scopus, 0 in GS.**

Conference Contributions

[OC1] P. Veselý, J. Minář, **A. Pražanová**, O. Šefl, and K. Dušek, "Novel Electrical Insulation Materials - Mechanical Performance of 3D Printed Polylactic Acid," *Proceedings of the 2020 International Conference on Diagnostics in Electrical Engineering, Diagnostika 2020*, Sep. 2020. DOI: 10.1109/DIAGNOSTIKA49114.2020.9214627,

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[OC2] **A. Pražanová** and V. Knap, "Electrical Circuit Model of Lithium-ion Batteries and Revisiting of its Parametrization Procedures," *ECS Transactions*, vol. 105, pp. 487–499, 1 Nov. 2021, ISSN: 1938-5862. DOI: 10.1149/10501.0487ECST/XML, **Contributions: AP: 50%, VK: 50%, citations: N/A in WoS, 1 in Scopus, 1 in GS.**

[OC3] V. Knap, **A. Pražanová**, and D.-I. Stroe, "Offline State-of-Health Estimation Method for Lithium-Ion Batteries in LEO CubeSats," in *43. Nekonvenční zdroje elektrické energie*, Česká elektrotechnická společnost, 2022, pp. 61–64, **Contributions: VK: 33.33%, AP: 33.33%, DIS: 33.33%, citations: N/A in WoS, N/A in Scopus, N/A in GS.**

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[OC5] V. Knap, **A. Pražanová**, J. Kasper, T. Finsterle, and P. Hrzina, "State-of-Charge Estimation Based on Open-Circuit Voltage Model Considering Hysteresis," *Proceedings of the International Spring Seminar on Electronics Technology*, 2024, ISSN: 21612536. DOI: 10.1109/ISSE61612.2024.10603756, **Contributions: VK: 20%, AP: 20%, JK: 20%, TF: 20%, PH: 20%, citations: 0 in WoS, 0 in Scopus, 0 in GS.**

[OC6] M. Fridrich, **A. Pražanová**, V. Knap, and J. Weinzettel, "Hydrogen in Automotive: LCA Study," *Proceedings of the International Spring Seminar on Electronics Technology*, 2024, ISSN: 21612536. DOI: 10.1109/ISSE61612.2024.10603647, **Contributions: MF: 25%, AP: 25%, VK: 25%, JW: 25%, citations: 2 in WoS, 2 in Scopus, 2 in GS.**

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Appendix A

Full Texts of the Core Publications

A. Pražanová, V. Knap, and D.-I. Stroe, "Literature Review, Recycling of Lithium-Ion Batteries from Electric Vehicles, Part I: Recycling Technology" [J1], © 2022 by the authors. This article is distributed under the terms and conditions of the CC BY 4.0 license.

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Review

Literature Review, Recycling of Lithium-Ion Batteries from Electric Vehicles, Part I: Recycling Technology

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Abstract: During recent years, emissions reduction has been tightened worldwide. Therefore, there is an increasing demand for electric vehicles (EVs) that can meet emission requirements. The growing number of new EVs increases the consumption of raw materials during production. Simultaneously, the number of used EVs and subsequently retired lithium-ion batteries (LIBs) that need to be disposed of is also increasing. According to the current approaches, the recycling process technology appears to be one of the most promising solutions for the End-of-Life (EOL) LIBs—recycling and reusing of waste materials would reduce raw materials production and environmental burden. According to this performed literature review, 263 publications about "Recycling of Lithium-ion Batteries from Electric Vehicles" were classified into five sections: Recycling Processes, Battery Composition, Environmental Impact, Economic Evaluation, and Recycling & Rest. The whole work reviews the current-state of publications dedicated to recycling LIBs from EVs in the techno-environmental-economic summary. This paper covers the first part of the review work; it is devoted to the recycling technology processes and points out the main study fields in recycling that were found during this work.

Keywords: lithium-ion battery; electric vehicles; recycling processes; technology of processes; battery recycling; battery reuse; electric vehicles; literature review

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1. Introduction

The ever-increasing number of requirements placed on decarbonization in the transportation sector result in an ever-increasing demand for electric vehicles (EVs) [1,2]. Lithium-ion batteries (LIBs) are the dominant energy storage technology for powering these vehicles due to their high volumetric and gravimetric energy density, long calendar life, low maintenance, and high-power capability. Thus, LIBs can meet the demands of the user in terms of high torque and speed, as well as range [3]. In the coming decade, the demand for EVs is expected to further increase; thus, the production on the lithium-ion market is predicted to grow exponentially [2]. Consequently, the demand for raw material will rise [4], while concerns regarding the availability of critical materials, in particular lithium (Li) and cobalt (Co), will further increase [5].

Therefore, ways for reducing the consumption of raw materials are being sought. Although their reuse in secondary applications extends the battery lifespan and increases benefits of their usage, one of the most promising ways for handling the End-of-Life (EOL) LIBs seems to be recycling [6–9]. Recycling allows for the recovering of valuable metals, securing the alternative materials supply chain, or gaining independence from exporting economies [10]. Therefore, all parameters of the techno-environmental-economic character are affected.

Recent studies describe that the recycling processes of retired automotive LIBs usually start with the pretreatment method, including discharging, dismantling, and any form of

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treatment (chemical, thermal) [11–13]. The recycling processes can be classified into (i) metallurgical/mechanical methods [14,15], including industrially expanded pyrometallurgical and hydrometallurgical technology, and (ii) laboratory examined direct recycling [13,16–19]. Both approaches, the industrial- and the laboratory-driven recycling, complement each other to achieve the highest possible efficiency of material recovery [20]. However, the recycling process carries an environmental burden, and in addition, its implementation is very costly [3].

This paper is the first part of a literature review study of peer-reviewed articles that discuss the "Recycling of Lithium-ion Batteries from Electric Vehicles" from a techno-environmental-economic perspective. In total, 263 publications have been summarized in the total work and divided into five sections: Recycling Processes, Battery Composition, Environmental Impact, Economic Evaluation, and Recycling & Rest. In this part of review, the technological point of view described in 89 publications devoted to recycling processes was summarized. The motive of this work is to create an overview mapping the current state of recycling processes of the LIBs from EVs (more than 96% of publications have been published in the last decade). Another review will be devoted to the environmental and economic perspective; all parts together will provide a comprehensive techno-environmental-economic analysis of this field.

The remainder of this paper is structured as follows: in Section 2 the methodology of the performed literature review is described, and Section 3 summarizes the current state of technology for the recycling processes of retired LIBs retired from EVs. Finally, Section 4 provides a conclusion and points to the main study fields identified by this review in the technology of procedures.

2. Review Methodology

The literature review was completed during September and October 2021 using two main databases, Web of Science (WoS) and Scopus, focusing on the variation of terms "recycling", "Lithium-ion", and "electric-vehicles". Articles, reviews, proceedings papers, early accesses, editorial materials, and letters were identified using the following title, abstract, and keyword search terms respecting the searching profile of the database. No search requirement for the year of publication was specified for this literature review; nevertheless, more than 90% of the publications from this review have been published in the last 10 years.

The main search was performed on WoS, where the keywords were differentiated according to the searching fields TOPIC and TITLE. Search by TOPIC includes title, abstract, and author keywords. On the contrary, search by TITLE refers to the title of a journal article, proceedings paper, or book. During these searches, the abbreviated forms of terms were distinguished, such as "Lithium-ion" and "Li-ion" or "Electric Vehicles" and "EVs". Moreover, a double search was performed for each searched term; enclosing the words in quotation marks retrieves records that contain the exact entered phrase. The number of publications found for TOPIC and TITLE terms is shown in Figure 1.

More than 600 articles, which were found following the aforementioned search methodology, were excluded from this review, because they covered unrelated content. The highest numbers of articles were found for all the searching terms by using the TOPIC field. Nevertheless, using the searching by TITLE brings concrete results that match the sought-after terms.

During the Scopus search, only two terms were used for filtering: "recycling AND lithium-ion", and "recycling AND li-ion", combined using the Boolean AND, with 1681 and 504 articles found, respectively. This search was used as supplement to the main search, which was performed in WoS. If the articles were not selected using the WoS search and met the content criteria, they were added to the literature review.

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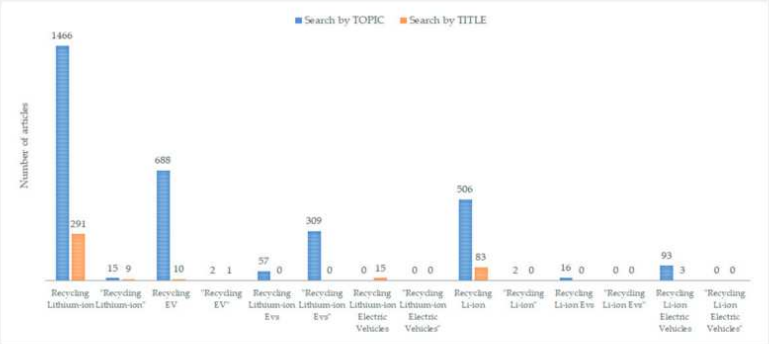

Figure 1. The main search on WoS, the number of searched articles of individual search terms.

The resulting publications were individually filtered by titles and clustered into the following five sections described by specific keywords: Recycling Processes, Battery Composition, Environmental Impact, Economical Evaluation, and Recycling & Rest. The keywords used for selection criteria and the descriptions of individual sections are summarised in Table 1.

Table 1. Keywords, the selection criteria for resulting publication of database search.

Section	Description	Keywords
Recycling Processes	Individual steps of the recycling process; description of special methods	Pretreatment Metallurgy/Mechanical Pyrometallurgy Hydrometallurgy Direct Recycling Special Method
Battery Composition	Characterization of recycling and recovery processes of the individual battery components	Cathode Anode Electrolyte
Environmental Impact	Issues related to the environment: general impact, Life Cycle Assessment (LCA) studies, focus on raw/reused materials	Envi/Ecological Impact Life Cycle Assessment (LCA) Recovery of Materials Recycling of Materials
Economic Assessment	Various types of economic evaluation, mainly including cost-benefit analysis	Economic Assessment
Recycling & Rest	Studies describing: the whole recycling cycle or approach it in a specific way; focus on EVs	Recycling EV LIBs Recycling LIBs

The five selected sections were arranged to completely cover the entire techno-environ mental-economic impact of recycling EV-retired LIBs. Because the individual parts of this investigation are intertwined, there is an overlap in the techno-environmental-economic scope of the resulting sections that is captured by Table 2.

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Table 2. Scope of the selected sections.

Section	Technological	Environmental	Economical
Recycling Processes	x		
Battery Composition	x	x	
Environmental Impact		x	
Economic Assessment			x
Recycling & Rest	x	x	x

All the publications were one-by-one reviewed for exclusion based on the same selection criteria as shown in Table 1. In total, 263 publications corresponding to the requirements of this literature review were selected. From these, 89 publications were analyzed according to the main topic of this paper and are listed in Appendix A. The current state of research & development (R&D) in focus on the technology of recycling processes of LIBs from EVs was discussed, and the main directions identified in this field of study were pointed out.

3. Results

All the selected 263 publications were divided into sections according to the methodology described in Section 2. Many of these papers thematically fall into several categories. The exact number of publications per category (labeled as a keyword), the percentage in the whole literature review, and their overlap are summarized in Table 3.

Table 3. Distribution of publications, the methodology of performed literature review.

Section	No. of Articles in Section	% of Total LR	Category/Keyword	No. of Articles in Category	No. of Overlapped Articles
Recycling Processes	61	23	Pretreatment	3	0
			Metallurgy/Mechanical	6	1
			Pyrometallurgy	4	0
			Hydrometallurgy	19	11
			Direct Recycling	6	3
			Special Method	23	9
Battery Composition	63	24	Cathode	51	51
			Anode	10	10
			Electrolyte	2	2
Environmental Impact	76	28	Envi/Ecological Impact	10	2
			Life Cycle Assessment (LCA)	8	3
			Recovery of Materials	35	1
			Recycling of Materials	23	0
Economical Assessment	15	6	Economical Evaluation	15	3
Recycling & Rest	48	18	Recycling EV LIBs	11	6
			Recycling LIBs	37	19

In total, 89 papers were selected from these publications (all sections) that characterized as the "Recycling Technology". The representation and overlapping in the individual categories are shown in Figure 2. In this figure, a connection between the categories is visible. For example, the hydrometallurgical processes are also linked to direct recycling (6), special methods (4), recycling retired automotive LIBs (3), and active cathode materials

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(2). Only one publication was found that was devoted directly to electrolyte processing in this literature review.

Of the selected 89 publications about the technology of recycling processes of LIBs from EVS, there were 53 Articles, 31 Reviews, 4 Proceeding Papers, and 1 Editorial Material. The distribution according to the type of publication in the individual category related to the technology of the processes is shown in Figure 3.

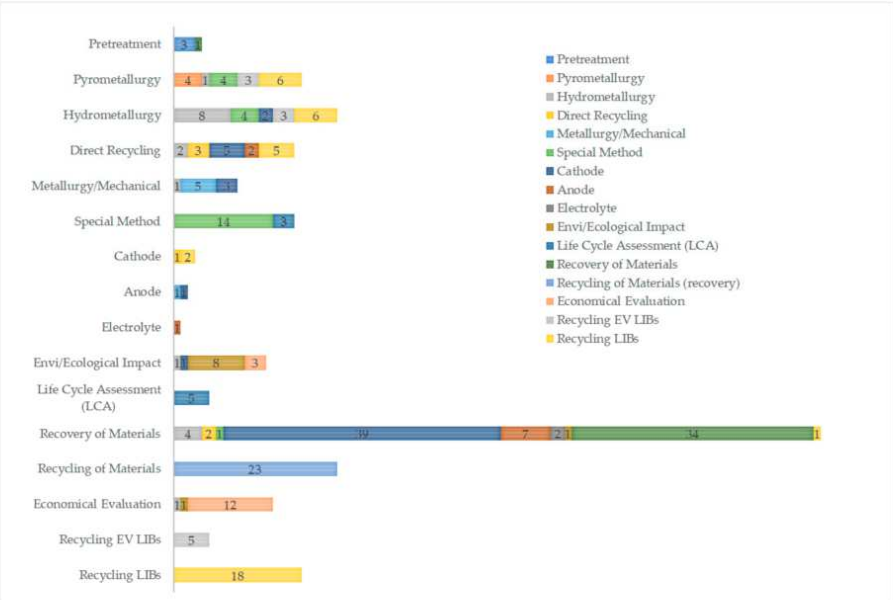

Figure 2. Total distribution and content overlap for publication selected according to the literature review in categories.

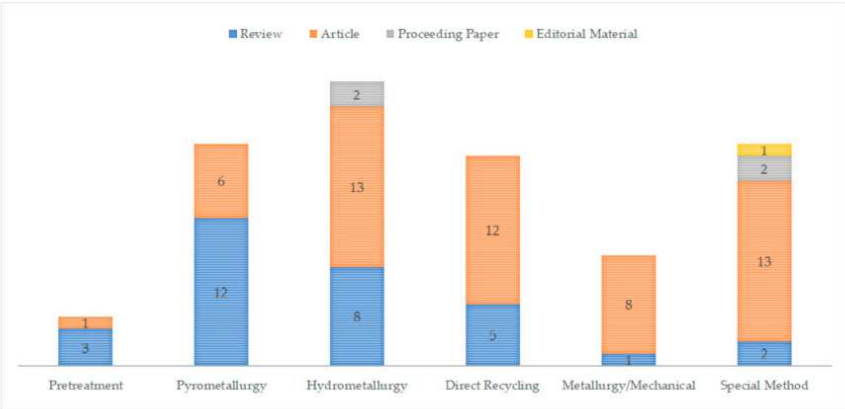

Figure 3. Distribution of “Recycling Technology” publications according to their type.

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3.1. Pretreatment of the Recycling Processes

The techniques of the recycling pretreatment process were described in a total of 4 publications according to the performed literature research as shown in Figures 2 and 3; there are 2 reviews and 1 article in the "Pretreatment" category, and 1 review in "Recovery of Material" one. These papers are summarized in Table A1 of Appendix A, containing the type, publication year, and a keyword summary of the primary content.

The pretreatment recycling techniques for lithium-ion batteries can improve the efficiency of recovering the valuable materials from the LIBs and lower the energy consumption in the following processes. These processes are used for the separation of the cathode materials from the battery casing, separator, current collector, electrolyte, additives, and connections. Approaches to pretreatment are different, but generally can be divided into experimental/laboratory-scale or large/industrial-scale methods. Laboratory-scale methods, which excel in the separation of active materials and process efficiency, are mainly focused on leaching and/or subsequent metal recovery. The industrial-scale processes are higher in process capacity and throughput, but separation of metals is less refined [11,13].

There is no well-defined designation for each pretreatment process category; thus, many variations have been used in recent years. The previously used division into mechanical separation, mechanochemical processes, thermal treatment, and dissolution processes [21,22] can be described in more apt categories yet. The usual three-step division of battery into discharging, dismantling, and separation (corresponding to laboratory-scale methods) was in [11] extended to a seven-step process: discharging, dismantling, comminution, classification, separation, dissolution, and thermal treatment, that fully captures the techniques applied in the industrial-scale processes.

3.1.1. Laboratory-Scale Pretreatment Methods

The most precise laboratory preparation process is described by a three-step method [11,13] usually consisting of:

1. Discharging

The lithium-ion battery cell is discharged by soaking in a saturated brine solution for 24 h. This process reduces the generation of short-circuiting and exothermic reactions of material deposits in the anode. For example, the response of lithium (Li) with oxygen and water leads to the inflammation of the highly combustible organic solvent [13].

2. Dismantling

The battery casing is manually disassembly to regain the cathode active material and the aluminum (Al) current collectors [13].

3. Separation

For cathode active material separation from the current collectors, binding reagents, and conductive additives, one of the following procedures can be used:

- High-temperature calcination is carried out between 350 °C and 600 °C to decompose the organic binders, additives, and electrolyte and release the active material in a powder form;
- Dissolution of polyvinylidene fluoride (PVDF) in N-methyl-2-pyrrolidone (NMP) supported using heat and/or sonication, subsequently following processes of drying and filtration;
- Dissolving the Al current collector using a strong base [13], e.g., sodium hydroxide NaOH [23].

NMP dissolution is nonpolar; thus, it is not applied on electrodes that contain a poly (tetrafluoroethylene)-based binder. Instead, a polar PVDF is used [13].

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3.1.2. Industrial-Scale Pretreatment Methods

The classification, presented in [11], summarizes in detail the industrial pretreatment, which is divided into seven steps and is illustrated in Figure 4. Every step of this approach fully respects the initial stage of following recycling processes.

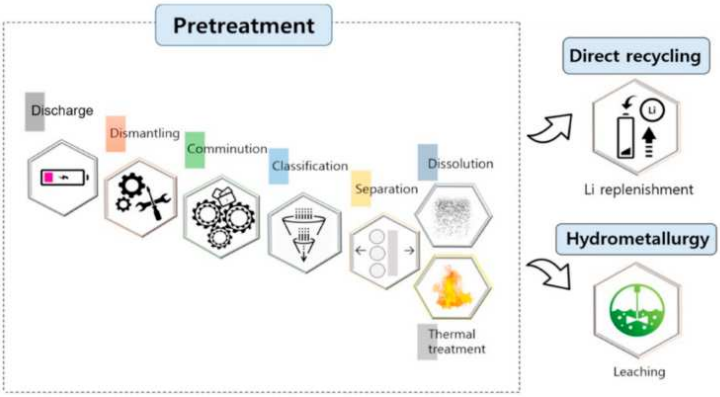

Figure 4. The scope and the sequence of the pretreatment process in LIB recycling [11]. Reprinted from Journal of Cleaner Production, Vol. 294, S. Kim et al., A comprehensive review on the pretreatment process in lithium-ion battery recycling, no. 8, pp. 1–29, Copyright (2021), with permission from Elsevier.

1. Discharging

The retired LIBs are in industrial processes treated variously. However, the discharging, as in laboratory-scale methods [13] is carried out by soaking for 24 h in one of the solutions:

- Distilled water;
- Sodium chloride (NaCl) water-based solution (using 10 wt% NaCl solution ensures the best discharging conditions; the extraction of the valuable metals is maximal);
- Alternative research-focused solutions such as potassium chloride (KCl), sodium nitrate ($NaNO_3$), manganese(II) sulfate ($MnSO_4$), magnesium sulfate ($MgSO_4$), and iron(II) sulfate ($FeSO_4$) [11].

2. Dismantling (disassembly)

The individual retired cells are usually manually dismantled or disassembled with knives and saws. These techniques adopt simple protective measures to fulfil the safety requirements [11].

There is one non-destructive automatic disassembly methodology used to recycle pouch-type LIBs, named Z-folded electrode-separator compounds. Using automatically stretching the Z-folded separators, the cathode and the anode are scraped off by the suitable toolset [11,23].

3. Comminution (mechanical treatment)

The dismantled LIBs need crushing and grinding to release the materials of the electrodes. The comminution step is essential for the recycling of the hydrometallurgical process [11].

The mechanical techniques differ according to the required particle size for further processing. Comminution can be mainly divided into [11]:

- Dry processes—crushing is conducted in a gastight unit in an inert atmosphere, generally in a two step-method in a low-speed rotary mill and high-speed impact mill [23], or a combination of the hammer crushing combined with a two-blade rotor crusher that can maximize the efficiency of this process [11];

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- Wet processes—the comminution equipment is a blade crusher with a water-based medium; firstly, the batteries are cut into pieces in a shear crusher, and the outputs are then crushed using the impact crusher. Then, water feeds into an entrance of the crusher and the particles in the form of a slurry carry the broken fractions through a selective sieve [11,23].

To enhance the efficiency of leaching of materials (extractions of Li and Co) in the processes, a mechanochemical reduction using planetary ball mill, attrition scrubbing, or grinding flotation is often used. The repetition of the crushing procedure increases the yield of the black mass, which is the shredded materials consisting of high amounts of lithium, cobalt, manganese, and nickel metals [11,24].

4. Classification (sieving)

According to the size of produced fragments from the LIBs, the material composition of obtained products can be determined. For example, products smaller than 850 µm consisted of the Lithium Cobalt Dioxide (LCO) particles and carbon, whilst materials as plastic, steel, aluminum (Al), and copper (Cu) formed enlarged compounds [11].

The particle size also affects the overall efficiency of the recycling process [11], where, e.g., a 500 °C optimum pyrolysis temperature was determined according to the maximum amount of the 0.2 mm size fraction in the pyrolytic cathode [25]; material extraction of the hydrometallurgical process was the most Fe (65%) was contained in the particles cruder than 6.7 mm, the particles of Al and Cu were mainly observed for the size of 2 and 6.7 mm, and the electrode elements, including Li, Co, manganese (Mn), and nickel (Ni), set from 92 to 98% in the particles finer than 6.7 mm [26].

5. Separation

The crude separation depends on the size of particles; more purified separation techniques, such as magnetic, eddy current, electrostatic, gravity separation, and froth flotation, ultrasonication, agitation, air separation, and density separation being used. The first three mentioned processes are described below, while the rest of the methods are listed in detail in the review [11]. Different approaches for the separation process are provided in [11,12].

- The magnetic separation removes the iron (Fe)-containing components and separates the cathode that contains active materials, the Al current collector, the anode, the steel casings, and the packaging [27].
- The eddy current separates the electrical conductors from the non-conductors or the minimally conductive materials. This method provides a high-ranking separation between Al and Cu in the electromagnetic fraction and Co and Li in the non-electromagnetic fragments [28].
- The differences in material electrical properties are utilized in the electrostatic separation. When an electric field is applied, charged or polarized particles are being moved and sorted from the LIBs crushed mass [29].

6. Dissolution

Even at this stage of the process after the classification and separation stage, some active materials are still fixed to the current collectors. The binders connect the other detached active materials.

For the separation of the cathode, dissolution of the binders, and the Al foils, N-methyl pyrrolidone (NMP) is typically used—the best solvent for polyvinylidene fluoride (PVDF), or, e.g., N,N-dimethylacetamide (DMAC), N,N-dimethylformamide (DMF), NeN- dimethyl sulfoxide (DMSO), ethanol, or molten salt of $AlCl_3$-NaCl [11].

7. Thermal treatment

At this stage, the binder materials that bring together the active materials and carbon conductive agents are still fixed to the current collectors [11]. For removal, some of the following thermal treatments can be used:

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- A two-step thermal treatment followed by calcination. The furnace temperature range varies from process to process and affects the overall duration. The first thermal step can be conducted between 150–500 °C and lasts for 1–2 h; the second is between 500–900 °C and lasts for the same length [30,31]. The calcination at 600–700 °C lasts about 5 h [11].
- The vacuum pyrolysis, which is carried out in a vacuum furnace during temperatures 500–600 °C [32]. The cathode materials are directly placed into the furnace to reduce the material loss [11].

The pyrolysis method can be combined, e.g., with ultrasonication or microwave methods [11].

3.2. Recycling Processes

The recycling processes were reported in 85 publications according to the literature search; the total number of publications is given by the sum of papers in individual categories: 9 in the "Metallurgy/Mechanical", 18 publications in the "Pyrometallurgy", 23 in the "Hydrometallurgy", 17 in the "Direct Recycling", and 18 in the "Special Method". These papers are described in more detail in relevant Tables A2–A6 of each category in Appendix A that contains the type, publication year, and the keyword summary of the primary content.

Overall, the recycling process brings an advantage in material savings (especially for valuable metals) and has a positive effect on energy consumption and the protection of the environment. Material production needs high energy for virgin resources' extraction and causes the release of greenhouse gases (GHGs) from transportation and smelting processes. Both the energy consumption and the amount of released GHGs can be reduced by reusing and recycling [13,33].

Recycling methods are generally used in industry, but there is still a lot of ongoing research carried out on the experimental/laboratory-scale to make recycling processes more efficient [33]. In Table 4, lithium-ion battery recycling facilities worldwide are listed according to [13,33–35]. For all companies, their location and their pretreatment and/or recycling process are described. The main products presented are for European facilities. For example, the German company Accurec developed a facility which recycles almost all the components from LIBs. Firstly, the batteries are disassembled into components, electronic parts, cables, and plastics; next, the remaining cells are sent to pyrolysis carried out at 250 °C. The subsequent mechanical processes separate ferromagnetic steel, aluminum cases, and Al and Cu foil from electrodes. The vacuum pyrolysis recovers lithium in the pure metallic form by evaporation and distillation or in the form of lithium oxide using selective gas evaporation of Li [18,36].

The recycling process from the French company Recupyl can be characterized by mechanical processing under an inert atmosphere of carbon dioxide (CO_2) and further hydrometallurgical recycling. LIBs are classified and crushed in the atmosphere containing a mix of CO_2 and argon (Ar) in an inert chamber. The mechanical crushing is two-step using a rotary mill at less than 11 RPM and an impact mill at less than 90 RPM. Meanwhile, the water neutralizes the off gas of these processes. Large-size fractions such as metal cases, paper, plastics, and foils are sorted out from electrode materials by the mill. The high induction magnetic separator splits up steel pieces. The rest of the fraction represents non-magnetic materials, which are segregated according to their densities. Separated active minerals are leached (hydrometallurgical processed) in heavily stirred water under an atmosphere with low oxygen levels, from where the Li, Co, and Mn are recovered. This process is very similar to the one used by the Swiss company BatRec [13,37].

As stated in Table 4, the most-used industrial recycling techniques include metallurgical and mechanical procedures such as the pyrometallurgical process and the hydrometallurgical recycling method. Moreover, the direct recycling process is under intensive research. These methods have been detailed described in this review in Sections 3.2.1–3.2.5. Section 3.2.5 is devoted to presenting unique methods and techniques of recycling.

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Table 4. Major LIBs recycling facilities worldwide according to [13,33,35].

Company	Country	Process	Products
Accurec	Germany	Mechanical, electric furnace	Co alloy, Li_2Co_3
Albemarle	USA	Hydrometallurgical	-
AkkureSer + Boliden	Finland	Mechanical (AS), Copper refining (Boliden)	Black Mass
Battery Resourcers	USA	Hydrometallurgycal	-
BatRec	Switzerland	Mechanical, pyrometallurgycal	Material fractures
Brunp	China	Hydrometallurgycal	-
Duesenfeld	Germany	Mechanical, hydrometallurgycal	Co, Ni, Mn (active mat.), electrolyte
Eramet	France	Pyrometallurgy	Ferro-Ni/Ferro-Mn alloy
Farasis Energy	USA	Mechanical	-
GEM	China	Hydrometallurgycal	-
GHTECH	China	Hydrometallurgycal	-
Inmetco	USA	Pyrometallurgycal	-
Highpower International	China	Hydrometallurgycal	-
Neometals	Austria	Mechanical, hydrometallurgycal	Co, Ni, Cu, Li, Gr (less quality)
OnTo Technology	USA	Mechanical	-
Recupyl	France	Mechanical, hydrometallurgycal	Mn, Co, Li, Ni (less quality)
Redux	Germany, Austria	Mechanical, hydrometallurgycal	Co, Ni, Cu, Li, Gr (less quality)
Retriev	Canada, USA	Mechanical, hydrometallurgycal	-
SNAM	France	Pyrometallurgy	Black mass, Co, Cu, Ni
Sony/Sumitomo	Japan	Pyrometallurgycal	-
SungEel HiTech	South Korea	Hydrometallurgycal	-
TES-AMM	Singapore	Hydrometallurgycal	-
Umicore	Belgium	Pyrometallurgy, hydrometallurgy	Co, Ni, Cu (chemical form)
uRecycle	Sweden	Mechanical	Black mass

- means that information was not fund.

3.2.1. Metallurgical and Mechanical Processes

The metallurgical and mechanical process was described in a total of nine publications according to the review; there are four articles and one review in the "Metallurgical and Mechanical" category, three articles in the "Cathode", and one article in the "Hydrometallurgy". These papers are summarized in Table A2.

As outlined in the next sections, the recycling processes are created by long and complex process chains to accomplish the main recycling aims fulfilling the high environmental

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and economic requirements. In the past few years, various recycling procedures have been developed. These techniques can be divided into two general process routes: pretreatment of the processes (the mechanical treatment before the metallurgy) and a straight pyrometallurgy or hydrometallurgy technique, or their combination, e.g., the pyrometallurgy and subsequent hydrometallurgical treatment. Direct recycling is still in a phase of industrial implementation [10,21,33,38].

As described in Section 3.1.2, the LIBs can be pretreated mechanically, often before a thermal treatment. After the initial comminution, the materials and components are classified by the physical properties (particle size, form, density, and electric and magnetic properties). Afterward, the outputs are processed in further metallurgical treatments. Typical outputs of these processes are Al/Cu fractions (conducting foils), non-ferrous metals (casings), ferrous metals (casings and screws), and a fraction called black mass (active electrode materials). The black mass can be either processed by pyrometallurgy methods or treated directly in hydrometallurgy procedures. Before hydrometallurgy, thermal pretreatment is required for the removal of organic components and concentration of the metal content [10,39].

Using the pyrometallurgical method, Co, Cu, and Ni alloys (metallic phase) or matte (sulfidic phase), Al, Mn, and Li slag (oxidic phase), and flying ash are produced [7,10,40]. These products can be further treated by hydrometallurgical procedure/direct recycling to recover the individual metals. In hydrometallurgy, Co, Li, Mn, Ni, and graphite can be recovered [10,38].

Nowadays, requirements on a combination of mechanical methods with pyrometallurgical and/or hydrometallurgical procedures targeted to processing of black mass increase and are under intensive research and development [24]. The reason is that the precious active electrode materials, such as Co, Li, Ni, Mn, and graphite, are deposited in the black mass. Additionally, conducting salts or elements such as Al, Cu, and Fe can be found there as well [10,33,41]. The concentration ranges of the black mass components produced from layered oxide chemistries are given in Table 5 according to [10].

Table 5. Concentration ranges of major black mass components produced from layered oxides chemistries, according to [10].

Elements	Content [wt %]
Aluminum (Al)	1–5
Cobalt (Co)	3–33
Copper (Cu)	1–3
Iron (Fe)	0.1–0.3
Lithium (Li)	3.5–4
Manganese (Mn)	3–11
Graphite	approx. 35
Fluor (F)	2–4
Oxide (O)	0.5–1

There are many approaches for the mechanical pretreatment of recycling LIBs. Thermal treatment can be applied before or after the mechanical methods or may be omitted altogether. The thermal treatment is typically formed by pyrolysis at around 500 °C to reduce the energy and eliminate the organic and halogenic content. The pyrolyzed LIBs can be safely mechanically processed without fire hazards [10,36]. In the other cases, specific safety measures are mandatory to prevent explosion and ignition during the processes. The non-thermal-treated procedures include for instance crushing under inert atmosphere of nitrogen (N_2), carbon dioxide (CO_2), or argon (Ar), followed by recovery of the volatile

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components by vacuum distillation or drying at moderate temperatures, or comminution in a solution, e.g., in a slightly alkaline medium [10,42].

Monitoring the effect of mechanical procedures has been investigated in many experiments [10]; for example, the influence of a second crushing on process and product was observed in [21]. In this experiment, a cutting mill with a discharge screen of 10 mm was used, and two different routes with/without the second crushing method were compared. Because of the comminution of the black mass fragments and the decomposition of inclusions, the second crushing increases the yield of black mass from 60% to 75%, as shown in Figure 5 from [39].

The black mass can be industrially processed by pyrometallurgical routes, described in Section 3.2.2, or treated by hydrometallurgical methods, described in Section 3.2.3. Due to the significant reduction of Al, other organic compounds, and excessive Co and Ni concentrations, the pyrometallurgy is preferred for black mass treatment. However, the F and Li content lead to corrosion, and the recovery efficiency of Li is not optimal [10,18,43]. On the contrary, Mn, graphite, and Li can be effectively recycled by hydrometallurgical processes. Nonetheless, there are still many issues of this procedure, such as F recovery, Mn recycling control, the production of graphite products, or the sensitiveness to organics compounds that easily contaminate process water [38,44].

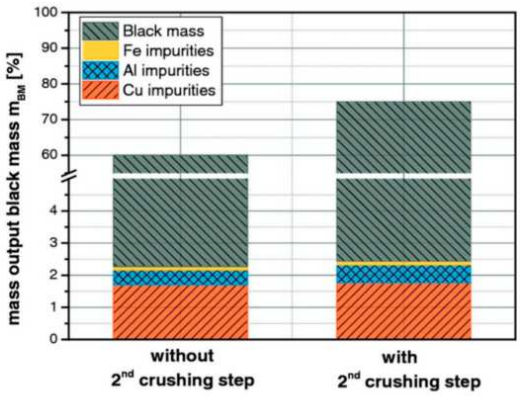

Figure 5. Yield of the black mass and impurities with/without 2nd crushing step [39]. Reprinted from Journal of the Electrochemical Society, Vol. 164, J. Diekmann et al., Ecological Recycling of Lithium-Ion Batteries from Electric Vehicles with Focus on Mechanical Processes, no. 1, pp. A6184–A6191, Copyright (2021), with permission from Journal of the Electrochemical Society.

3.2.2. Pyrometallurgical Process

The pyrometallurgical recycling technique was described in a total of 18 publications according to the performed search; there are 3 articles and 1 review in the "Pyrometallurgy" category, another 4 reviews and 2 articles in the "Recycling LIBs", 4 reviews in the "Special Method", 3 reviews in the "Recycling EV LIBs", and 1 article in the "Hydrometallurgy" category. These papers are summarized in Table A3.

The pyrometallurgy recycling technique uses a high-temperature furnace to reduce valuable metals and refine them through physical and chemical transformation. As temperature increases, at first structural changes, as phase transitions, occur. Later, at already high temperatures, the dominating process is chemical reactions, resulting in batteries being smelted. The required heat is typically provided by exothermic reactions, combustion, or electrical power, depending on various specifying factors such as temperature and processing time [9,43].

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Thermal pretreatment is significant for the pyrometallurgical process, and is described in detail in Section 3.1.2 [11]. Usually, these techniques are based on incineration, pyrolysis pretreatment, or other variations of these types [11,43].

The mainly used pyrometallurgical options conducted for recycling LIBs are roasting, calcination and smelting. These processes can be sorted according to the atmosphere, where the process is carried out and the extraction mechanisms used, as illustrated in Figure 6 [43].

Roasting is an exothermic mechanism, which at elevated heat involves reactions between gas and solid particles. Materials such as carbon, charcoal, or coke then act as a reducing agent, which heats up the cathode material, resulting in a mix of alloys and carbon residue. A lower valence state is then reached by the lithiated metal oxide. Additionally, further oxide reductions can happen by using the carbothermic reduction carbon at the required temperature [45,46]. The amount of metal oxide reduction in this process is then controlled by the quantity of carbon participating [43,45,46].

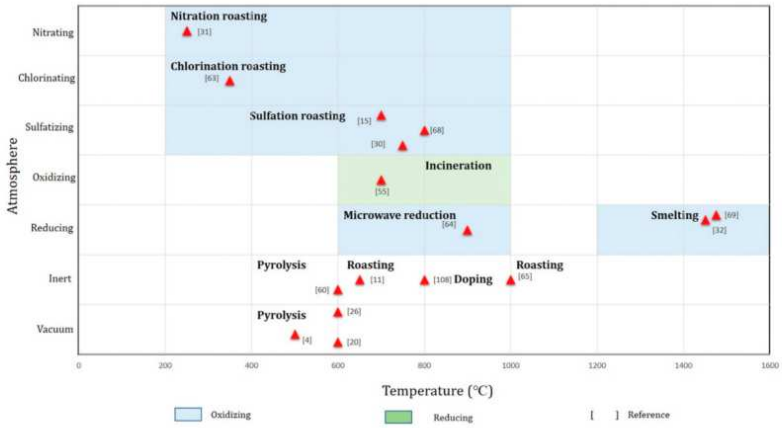

Figure 6. A brief data review of published work on pyrometallurgical options for recycling Li-ion batteries [43]. Reprinted from Journal of Power Sources, Vol. 491, B. Makuza et al., Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review, Copyright (2021), with permission from Elsevier.

The roasted mechanism was for example, realized by [43] where a mixture of Lithium Cobalt Dioxide (LCO) and graphite were treated under a nitrogen atmosphere for 30 min at a temperature of 1000 °C. The residue after roasting was water leached [43]. The recovery rates were 98.93% for Li, 95.72% for Co, and 91.05% for graphite [43,47]. In another study [48], a temperature of 800 °C was used to roast Lithium Manganese Dioxide (LMO) for 45 min. For the LMO reduction, graphite was used in the mix of lithium carbonate (Li_2CO_3) and manganese(II) oxide (MnO) subsequent with water-based leaching and mechanical separation. Recovery of this process was 99.13% Li and 95.11% manganese(II,III) oxide (Mn_3O_4) [43,48].

A change from carbothermic reduction roasting to salt-assisted reduction roasting is recently applied. The salt-assisted procedures can reduce costs by lowering the excessive energy needs for evaporative crystallization. Moreover, salt-assisted roasting can presumably increase the efficiency of the whole recycling process, reducing acid consumption and toxic gas emissions by the production of water-soluble salts. According to the used reagent, other roasting procedures, such as chlorination, sulfation, and nitration, can be classified [43].

The smelting process allows the recycling of various EOL LIBs based on different chemistries [9]. By smelting, the material of the retired battery is heated above its melting

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point and subsequently the metals are separate in the liquid phase by reduction and subsequent formation of immiscible molten layers [43,49]. Moreover, this process eliminates the necessity for a previous passivation step, and the battery cells can be directly thrown into the smelting furnace. The process of smelting usually consists of two complementary phases [43]:

- At first, the battery material is heated at a lower temperature to slowly evaporate the electrolyte [14];
- Secondly, the temperature is increased to melt the material's feeds [43].

The organic material is burnt out to provide energy for the process. The reduction of the active cathode material is regulated in a blast or electric furnace. After the smelting, the transition metals are concentrated into a molten alloy phase. For the recovery of valuable metals, the hydrometallurgical technique is consequently used [34,43].

Pyrometallurgical recycling techniques have many advantages, such as that the batteries do not have to be pre-treated (e.g., before smelting), and the material recovery yields are high (more than 90%) [43]. However, this process is costly [48,50]. Further advantages and disadvantages of this process are listed according to [51] in Table 6.

Table 6. Advantages and disadvantages of pyrometallurgical process according to [51].

Advantages	Disadvantages
Application flexibility; all battery compositions, and configurations	Not possible to obtain products based on: Li, Al, organic materials
Not required pretreatment (sorting, mechanical processing)	Unable to recycle Lithium Iron Phosphate (LFP)
High proportion of recovery metals in products	High energy and capacity requirements
Proven technology; existing equipment can be used	Expensive gas cleaning; prevention of toxic emissions in the air

3.2.3. Hydrometallurgical Process

The hydrometallurgical recycling method was described in a total of 23 publications according to literature review; there are 7 articles and 1 review in the "Hydrometallurgy" category, 4 reviews and 2 articles in the "Recycling LIBs", 3 articles and 1 review in the "Special Method", 1 review and 2 proceeding papers in the "Recycling EV LIBs", and 1 article and 1 review in the "Cathode" category. These papers are summarized in Table A4.

Various hydrometallurgy techniques were developed in recent times for recycling cathode active materials of different chemistries LIBs including Lithium Cobalt Dioxide (LCO), Lithium Manganese Dioxide (LMO), Lithium Nickel Manganese Cobalt Oxide, (NMC), Lithium Nickel Cobalt Aluminum Oxide (NCA), and Lithium Iron Phosphate (LFP) [52], to recover valuable metals such as Co, Ni, Mn, and Li [53,54]. The whole hydrometallurgical process is created by physical and chemical methods proceeding in liquid media, which allow the high recovery of these metals [26,55,56].

The physical operations include the mechanical pretreatment methods such as sieving and magnetic separation described in Section 3.1.2. [11]. The LIBs are discharged and separately treated to improve the safety and recovery rate of the whole process [55,57]. This operation allows the conservation of the minerals and efficiently separates the individual metals [55].

The chemical operations can be classified according to the procedures and final products [46] as illustrated in Figure 7 from review [58].

- The first stage, "Leaching", contains dissolving or leaching of the valuable metals by acid or basic agent in an oxidizing or reducing medium in leaching tanks [55,57]. Many inorganic acids such as hydrochloric acid (HCl), sulfuric acid (H_2SO_4), hydrogen

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peroxide (H_2O_2), and nitric acid (HNO_3) [59] or organic acids, e.g., citric acid, malic acid, oxalic acid, etc., are usually used [60,61].

- The second stage "Impurity removal" is created by solid-liquid separation, which clarifies the leach solution by filtration or centrifugation [55,62].
- The last stage "Ni, Co, Mn, and Li recovery" is devoted to the final recovery of valuable metals in hydroxide or metal salts [55]. This process includes, for example, solvent extraction [63], electrochemical techniques [64], selective precipitation [65], and separation by ion exchange resins [55].

Different leaching techniques are used for various battery chemical compositions to achieve the most efficient recovery of materials. The most promising current state treatment reagents belong to hydrochloric acid (HCl), used for LCO batteries [52,66–69]. According to the experimental results of [66–68], the leaching efficiency of valuable metals (Co and Li) is achieved 100% for 2 M and 4 M HCl at a temperature of 80 °C in 90 min and 60 min-long extraction, respectively. In other experiment, the leaching efficiency was higher than 99% of all materials (Co, Ni, Mn, and Li) by using 4 M HCl at 80 °C for 60 min [69].

Many other studies ([52,70,71]) were to the leaching reactions of nitric acid (HNO_3) devoted. For example the 100% recovery of Li and 95% recovery of Mn was achieved by leaching using 2 M HNO_3 at 80 °C within 120 min [70]. The combination of 1 M HNO_3 and 1.7 vol% hydrogen peroxide (H_2O_2) leached at 75 °C for 30 min increased the recovery of Co and Li up to 99% [71].

Leaching sulfuric acid (H_2SO_4) procedures are usually slow and the metal recovery efficiency is lower. By using 2 M H_2SO_4 at 70 °C, just 76% efficiency was observed [71]. When combining H_2SO_4 with H_2O_2, the experiment efficiency is approximately equal to the values observed in HNO_3 experiments. The optimum concentration according to the ref. [52] is 2 M HNO_3 and 5 vol% H_2O_2 for 60–90 min long leaching time at temperature between 60–80 °C, and a solid/liquid ratio of 50 g L^{-1}.

A detailed description of other procedures using reagents such as oxalate, ammonia, DL-malic acid, phosphoric acid, etc., additionally supplemented with overview tables, is provided, for example, by reviews [13,52,72].

Hydrometallurgical recycling techniques have many advantages such as the high recycling process efficiency and pureness of the final products [51]. However, the LIBs must be subjected to both procedure steps of the hydrometallurgical method—physical pretreatment and chemical recovery [52]. Further advantages and disadvantages of this process are shown in Table 7 according to [51].

Table 7. Advantages and disadvantages of hydrometallurgical process according to [51].

Advantages	Disadvantages
Application flexibility; all battery compositions and configurations	Necessary to crush the batteries; high safety requirements
Flexibility of separation process; a desired product (metal) can be obtained	Uneconomical for Lithium Iron Phosphate (LFP)
High efficiency of the recycling process (especially for Li)	High volume of waste water; necessary disposal or further recycling
High purity of products	Impossibility of recycling anode materials (graphite, conductive additives)
Emission-free	High operating costs

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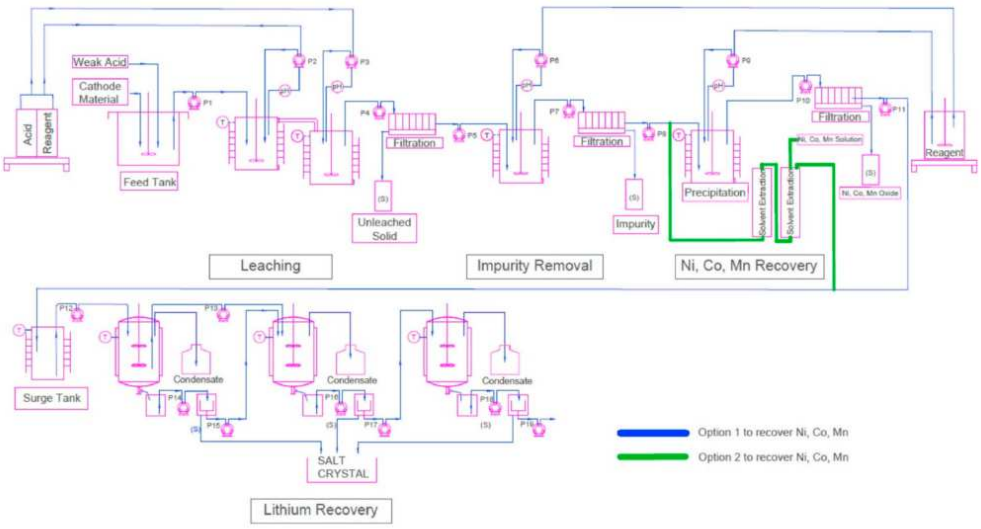

Figure 7. Schematic of the hydrometallurgy process for recycling battery cathode materials [58]. Reprinted from Journal of Energy Storage, Vol. 35, J. C. Y. Jung, P. C. Sui, and J. Zhang, A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments, Copyright (2021), with permission from Elsevier.

3.2.4. Direct Recycling Process

The process of direct recycling was totally described in 17 publications according to the performed review; there are 3 articles in the "Direct Recycling" category, 5 articles in the "Cathode", 1 article and 1 review in the "Anode", 1 article and 1 review in the "Hydrometallurgy", and 3 reviews and 2 articles in the "Recycling LIBs. These papers are summarized in Table A5.

The direct recycling process handles the total recovery of LIBs materials for reusing in a new battery production instead of dissolving the active material entirely by hydrometallurgical techniques [13,58]. This process consists of various physical and chemical steps with low temperature and energy requirements used for battery separation. Therefore, the procedure should be lower in costs than leaching because the intervention of the material is minimized [58]. Direct recycling is not commercialized yet [51,58]; there are only some published processes from recycling companies. Selected companies are listed in Table 4. Detailed summaries of direct recycling processes of individual companies are provided by reviews [58,73,74].

For instance, the process patented by company Retriev with a capacity recovery of 95% consists of crushing and screening mechanisms followed by thermal treatment at 500 °C for the carbon surface modification and liberation of polyvinylidene fluoride (PVDF) [75]. For carbon removal, selective flotation is implemented; the calcination at 500–800 °C is realized using a fraction mixture and lithium hydroxide solution [58,75]. Alternatively, the mechanism of the American facility of OnTo Technology is executed in the atmosphere of supercritical carbon dioxide (CO_2), as an alkaline solution is used with lithium hydroxide (LiOH), and the calcination is performed at 400–900 °C [58].

As shown in Tables 5 and 6, the pyrometallurgical and hydrometallurgical process are not optimal for recycling Lithium Iron Phosphate (LFP) batteries. Therefore, direct recycling research is aimed at their recycling [58].

The cathodes of the pretreated LFP batteries with sorted components were submitted to thermal reactivation processes at 500 °C under nitrogen flow [76]. These processes

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decompose the polyvinylidene fluoride (PVDF) binder and the active LFP material from other features. The recovered cathode material has a low electrochemical capacity versus Li^0 (reaching 136 mAh/g at 0.1 C) [76]. Consequently, this process was improved by [77,78] at the pilot experiment by insertion a Li precursor and a reducing gas during the thermal treatment; hence, the electrochemical capacity versus Li^0 exhibits higher values (145 mAh/g at 0.1 C). The process flow chart of selective leaching process is shown in Figure 8 [58]. Moreover, the process integrated a two-temperature-washing-step with dimethyl carbonate (DMC) to remove organic compounds before component separation, milling, and thermal treatment at 200 and 500 °C in a nitrogen (N_2) atmosphere [78]. Regenerated LFP provides the highest capacity versus Li^0 (150 mAh/g at 0.1 C), but Al contamination at 0.5% was observed and had to be minimized to prevent accumulation in the recycled material [58].

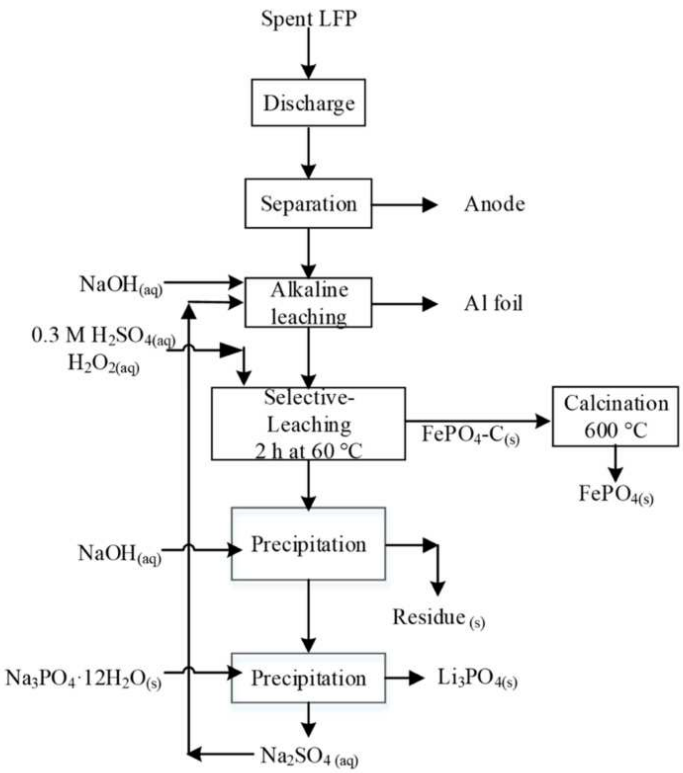

Figure 8. Selective leaching process [58]. Reprinted from Materials, Vol. 13, F. Larouche et al., Progress and Status of Hydrometallurgical and Direct Recycling of Li-Ion Batteries and Beyond, no. 3, p. 801, Copyright (2021), distributed under Creative Commons Attribution License.

Other direct recycling processes are being applied; for example, in a simple patented process by [79], very high-frequency ultrasound (>900 kHz) for up to 30 min was applied to the whole (not damaged) battery. Thereby the surface of the cathodes was cleaned out of absorbed phosphorous compounds that come from the dissociation of electrolyte lithium hexafluorophosphate (LiPF6) during cycling. Another leaching procedure was developed by [80], where the cathode materials are cleansed off residual Cu and Al in an alkaline solution (a complexing agent (5 M ammonium hydroxide NH_4OH) and 1 M

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lithium hydroxide (LiOH) with O_2 (g) bubbling to promote oxidation) after separation from other components. After 12 h, these contaminants are completely solubilized. Using the direct recycling procedure, any type of LIB can be treated. However, LFP batteries must comply with the specific pH requirements for process safety [58].

The direct recycling process is complicated in terms of pretreatment techniques that are necessary for battery material separation. On the other side, all the battery materials can be recycled with sufficient energy efficiency. Other advantages and disadvantages of the process of direct recycling are described according to [51] in Table 8.

Table 8. Advantages and disadvantages of the process of direct recycling according to [51].

Advantages	Disadvantages
Recycling of all materials: anodes, cathodes, electrolytes, foils, . . .	Difficult mechanical pretreatment, necessary material separation
Suitable for Lithium Iron Phosphate (LFP)	The mix of materials reduces the quality of the process
Energy efficient	Low quality of output products
Production residues can be recycled	Not yet fully industrially applied

3.2.5. Special Recycling Methods

The special methods describing recycling processes were characterized in a total of 18 publications according to the search; there are 11 articles, 2 reviews, 1 proceeding paper, and 1 editorial material in the "Special Method" category, and 2 articles and 1 proceeding paper in the "Life Cycle Assessment (LCA)". Many studies in these publications have been devoted to a precisely characterized process. Therefore, this essential content is summarized in Table A6.

The scope of these publications is diverse: refs. [81,82] deal with mathematical models; impacts of life cycle characterization of LIBs [83,84]; the environmental advantages and disadvantages discussion, e.g., [85,86]; industrial processes and projects description [87–91], and R&D projects in laboratory-scale definition [36,92–95].

The processes described in previous sections (Sections 3.2.2 and 3.2.3) have been summarized mainly at the industrial level; thus, a review gives a more detailed presentation of some studies from the experimental/laboratory-scale, not focusing only on direct recycling (Section 3.2.4), but supports the research of mentioned metallurgical procedures.

The pretreatment process before the recycling procedure in the laboratory-scale was described in detail, for example, in [11] or [13]. A review summarizing all the laboratory-scale recycling procedures was not found during this literature review. Only one article, published in 2001 [67], outlined "A laboratory-scale lithium-ion battery recycling process". This paper refers to experimental testing of commercial, cylindrical 18650 sizes $LiCoO_2/LiC_x$ retired batteries, which were treated by crushing and riddling to the extraction of the active materials. Next, the active materials were selectively separated using N-methyl pyrrolidone (NMP) at about 100 °C for 1 h; from the product, the Al and Cu foils, and lithium cobalt oxide ($LiCoO_2$) and carbon powder were filtrated. The dissolution of lithium cobalt oxide was achieved by treating the separated residual powders with a small volume of 4 M hydrochloric acid (HCl) for 1 h at about 80 °C, where the acid/sample ratio of cobalt and lithium from the oxide was 10. As a side product, carbon powder was removed using the solution decantation. The cobalt dissolved in the hydrochloric solution was recovered as cobalt hydroxide $Co(OH)_2$ by the addition of one equivalent volume of a 4 M sodium hydroxide (NaOH) solution. Consequently, the precipitation due to the $Co(OH)_2$ would be obtained for pH between 6–8, ensuring NaOH solution treatment [67].

Existing industrial techniques are currently being extended under a laboratory scale. For example, using molten salts within a temperature range of 800–900 °C as electrolytes and reaction media is being applied as a chemical recycling procedure. Different eutectic mixtures of molten salts such as sodium, potassium, lithium, and calcium borates and

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chlorides, sodium, and potassium carbonates are being tested to provide an optimized alternative to the hydrometallurgical or pyrometallurgical processes of metal recovery. The final metal purity for single-metal oxides for these processes is around 98–99% [92].

An innovative mechanochemical method for the cathode materials ($C/LiCoO_2$) of retired LIBs and waste polyvinyl chloride (PVC) was examined. The mixture of $LiCoO_2/PVC/Fe$ was co-grinded and further water leached. As a result of this procedure, recoverable lithium chloride (LiCl) from Li by the dechlorination of PVC and magnetic cobalt iron oxide ($CoFe_4O_6$) from Co were generated. This study can also be classified as environmentally friendly because it was found that the chlorine atoms in PVC are mechanochemically transformed into chloride ions that bond to the Li in lithium cobalt oxide ($LiCoO_2$) to form LiCl. This resulted in the reorganization of the Co and Fe crystals to create the magnetic material $CoFe_4O_6$. Therefore, including this recycling process in the existing chain would have both environmental and economic benefits [93].

4. Discussion and Conclusions

Electric vehicles are experiencing significant growth globally; hence, the need for Lithium-ion batteries is increasing as well. However, the best environmental and economic beneficial strategies of treating the retired LIBS are still being sought. Nowadays, the best scenario seems to be the combination of secondary applications of retired LIBs at the end of their primary lives in EVs (e.g., in stationary storage), followed by the recycling of End-of-Life (EOL) waste batteries [67,96,97].

In the last years, the recycling process of LIBs has been a much-discussed topic; the recycling procedure addresses the still-growing amount of EOL batteries and reduces the consumption of raw materials used in the new battery production. Recycling recovers valuable metals such as lithium (Li), cobalt (Co), nickel (Ni), and manganese (Mn) and other used materials, for example, aluminum (Al), copper (Cu), or iron (Fe); thus, it secures alternative materials supply chains and gains independence from exporting economies. Therefore, it can be stated that the choice of technological process of recycling will have a strong environmental and economic effect [10,11,13].

Thus, this comprehensive literature review, based on 263 publications, about "Recycling of Lithium-ion Batteries from Electric Vehicles" was performed providing a technological-environmental-economic summary of the current state of this topic. The whole work was divided into two separate parts: (i) the technology of recycling processes and (ii) their environmental and economic effects. This paper describes in review only the first part, namely the technological point of view of recycling processes and industrially and laboratory used strategies. A techno-environmental-economy overlap of recycling EOL LIBs focusing on the summarized content of this part of the whole review is shown in Figure 9.

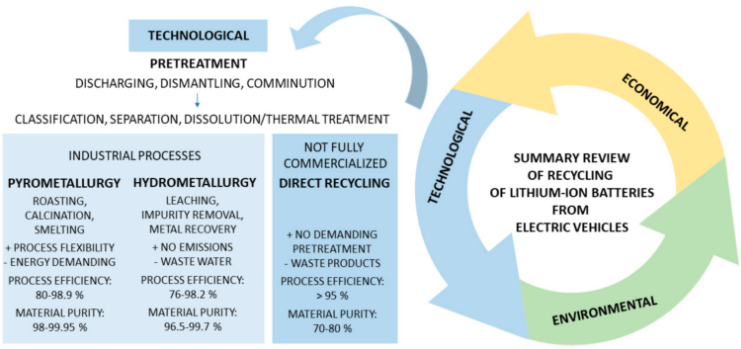

Figure 9. The summarizing figure of the first part of the literature review of recycling lithium-ion batteries from electric vehicles specialized on techniques of this process.

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All 89 articles devoted to recycling technology were reviewed according to their content and divided into six categories: pretreatment of the recycling processes; and recycling processes including metallurgical and mechanical processes, pyrometallurgical process, hydrometallurgical process, direct recycling, and special recycling methods. The summary content is listed in Tables A1–A6 of the Appendix A of this paper.

There is no well-defined designation for pretreatment processing of retired LIBs. The previously used four-step classification of pretreatment procedure appears to be insufficient for currently used industrial techniques. This process was best captured in [11], where the pretreatment was divided precisely, according to the methods and techniques, into the seven following steps: discharging, dismantling, comminution, classification, separation, dissolution, and thermal treatment. The intensity, methodology, and procedure choice of mechanical processing affect the quality and amount of output products (fractions of active materials called black mass). According to [39], the addition of the second crushing step into the mechanical pretreatment process increased the mass of black mass obtained by 15% compared to the single crushing process. Thus, the total process efficiency will be positively affected.

The pre-treated LIBs are subsequently metallurgically processed. The most industrially used techniques are pyro- and hydrometallurgy. These methods are in the right combination (pyrometallurgical method followed by the hydrometallurgical process) used for very effective recycling black mass, which can reach up to almost 100%. In the pyrometallurgical procedure, Co, Cu, and Ni alloys in the metallic or sulfidic phase, Al, Mn, and Li slag in oxidic phase, and flying ash can be produced. In the hydrometallurgical process, Co, Li, Mn, Ni, and graphite can be recovered [10,98].

The pyrometallurgy recycling reduction of valuable materials is based on the physical and chemical transformation of thermally treated LIBs. The lower temperature treatments secure structural changes, including phase transitions; the higher temperatures provide chemical reactions using roasting, calcination, or smelting. The efficiency of pyrometallurgical process in the recycling of electrode materials, LIB lithium slag, or scraps reached over 80–98.9%. The purity of recovered metals ranged between 98 and 99.95%. The hydrometallurgical technique requires only mechanical pretreatment of the discharged batteries, such as sieving and separation. Then, the batteries are chemically treated in liquid acids or basic agents, which allow a high recovery of metals. The efficiency of those processes is around 76–98.2%, the purity is set between 96.5–99.7%. When recycling anode materials by leaching, the overall efficiency was almost 100% with the resulting purity of the obtained products around 99.9% [17,38,98,99].

The direct recycling process represents another recycling technique. This procedure is created from many physical and chemical steps with low temperature and energy requirements used for battery separation. Thus, the procedure cost should be lower in comparison to hydrometallurgical leaching. Furthermore, the efficiency of the process still be higher than 95%, and the purity of recovered materials should be comparable to metallurgical processes. Direct recycling is not yet fully commercially available. The implementation is soon envisaged because treating this method allows for the complete recycling of LFP batteries, which is problematic using current processes. Nowadays, direct recycling is explained in procedures published by a few recycling companies or laboratories, which cooperate for full application [58,67].

Although the overall efficiency of the mentioned processes is mostly above 90%, their implementation is limited by their technological complexity, required output products, further processing, predicted, or already monitored environmental burden, and costs that can be provided for line equipment and operation. Even though the batteries can be pyrometallurgical recycled on existing facilities, the operating costs of the whole process are high (mainly because the process requires high temperatures). In addition, toxic gases and water are released. Pyrometallurgy does not allow the recycling of electrolyte or graphite from anode active materials. On the contrary, the equipment of the hydrometallurgical line is costly, but the operating costs are much lower (depending on the chemicals used). As an intermediate product,

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only toxic wastewater is discharged and is further treated for reuse. Even hydrometallurgy does not allow the recycling of electrolytes from batteries [33,100,101].

The currently used metallurgical processes have several disadvantages. For their optimal integration into the entire life cycle of LIBs, it would be necessary to reduce their environmental impact, whether in the form of generated emissions (pyrometallurgy) or wastewater (especially hydrometallurgy). These restrictions are complex and very expensive. Thus, the recycling integration could be reduced.

A solution that could encourage the expansion of recycling facilities is expected to implement the direct recycling process fully. The input costs for direct recycling line equipment are expected to be high. However, operating costs, waste intermediates and the environmental burden will be lower compared to the existing processes and therefore sustainable in the long-term run. Moreover, all types of LIBs can be recycled by direct recycling. The main shortcoming of the currently implemented direct recycling procedures is the resulting purity of the obtained materials, which ranges between 70–80%. The use of lower quality input products could fundamentally affect the characteristics of new batteries; thus, the effectiveness of metal recovery must be increased. This could be achieved by applying some steps of established metallurgical processes [33,102].

Research in the coming years shall focus on the complete application of the direct recycling process, increasing its metal recovery potential and lowering the total process costs. Within that, the optimization of current metallurgical processes would be desired because reducing the environmental burden could encourage further use of these recycling techniques.

Thereby, the main directions of current-state publications in the field of "Recycling of Lithium-ion Batteries from Electric Vehicles" are summarized in this review paper. The next work will be devoted to reviewing the rest of the publications, which are discussing the environmental and economic fields and provide answers to questions concerning environmental impact; comparison of Life Cycle Assessments (LCA) of LIBs; the contribution of recovery materials; and economic evaluation, mainly including cost-benefit analysis with consideration of extended lifetime in the secondary applications of retired LIBs.

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Appendix A

Table A1. Assessed literature in review for "Pretreatment" category.

Pretreatment of the Recycling Processes			
Reference	**Type**	**Publication Year**	**Summary Content**
[12]	Review	2021	Three-step pretreatment method, pretreating flowchart of retired LIBs
[11]	Review	2021	The innovative approach to the classification of pretreatment, seven-step method
[103]	Article	2020	Investigation of incineration of LIB cell materials
[13]	Review	2020	Procedures of lab-scale and industrial-scale pretreatment

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Table A2. Assessed literature in review for "Metallurgical and Mechanical Process" category.

Metallurgical and Mechanical Processes			
[14]	Article	2018	Two main basic aspects of recycling battery packs: mechanical procedure and chemical recycling (metallurgical)
[15]	Article	2017	Recycling Metals from LIBs; mechanical separation; vacuum metallurgy
[10]	Review	2020	The current status of development, focusing on the metallurgical processing of LIB modules and cells
[104]	Article	2017	In-situ recovery from retired LIBs; vacuum metallurgy
[39]	Article	2017	The LithoRec projects; energy-efficient recycling mechanical process-steps
[105]	Article	2019	Mechanical and hydrometallurgical processes in HCl media
[106]	Article	2020	Leaching of LNCM cathodes in ascorbic acid lixiviant
[77]	Article	2016	Decomposing of $LiFePO_4$ to host particles, recycling cathode powders using heat-treated at different temperatures
[61]	Article	2018	A "grave-to-cradle" process for the recycling of retired mixed-cathode materials

Table A3. Assessed literature in review for "Pyrometallurgical Process" category.

Pyrometallurgical Process			
Reference	**Type**	**Publication Year**	**Summary Content**
[107]	Article	2020	Erosion mechanism of refractories in a pyro-furnace
[43]	Review	2021	Overview on extractive pyrometallurgical options for recycling retired LIBs; lab-scale and industrial-scale processes
[72]	Review	2020	Overview on laboratory and industrial investigations and implementation of recycling
[24]	Article	2020	Recycling of pyrolyzed LIBs black mass, Li concentrates
[99]	Article	2021	Pyrometallurgical treating LCO in an Al_2O_3 and MgO crucible, concept for the treatment of LFP
[7]	Article	2020	Recycling processes; flowchart for pyrometallurgical recycling of Li-ion, Ni–Cd, and Ni–MH batteries.
[40]	Review	2018	A brief review of typical physical and chemical processes
[108]	Review	2021	Potential benefits of pretreatment; recycling processes
[21]	Review	2008	Structure of LIBs; overview of single and typical combined recycling processes;
[9]	Review	2019	Recycling process of EV LIBs
[17]	Review	2021	Reduction, reuse, and recycle (3R) of retired LIBs; pretreatment methods; technological processes
[48]	Review	2018	State-of-the-art research of recycling procedures; concept of suitability, LCA assessment of battery recycling
[109]	Article	2019	Quantitative analysis of the recycling methods
[100]	Review	2020	Improvements of recycling processes; a holistic design approach for LIBs
[18]	Review	2019	Overview of recycling commercial processes
[50]	Review	2018	Battery collection, transport, recycling commercial processes; End-of-Life (EOF) EV battery consideration
[110]	Article	2016	Recycling processes; hydrometallurgy for cathode recovery
[111]	Review	2014	Recycling procedures; problem and prospect analysis

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Table A4. Assessed literature in review for "Hydrometallurgical Process" category.

			Hydrometallurgical Process
[112]	Article	2019	Enhanced hydrometallurgical process; iron-precipitation, liquid-liquid extraction, innovative Li-Na separation
[62]	Article	2013	Overview of leaching and solvent extraction strategies
[44]	Article	2020	Experimental and modelling results for the hydrometallurgical recycling $LiCoO_2$ cathodes; physicochemical model
[113]	Article	2009	A hydrometallurgical route based on leaching-crystallization steps for the separation of metals Al, Co, Cu and Li
[114]	Article	2021	Recycling processes; evaluation of effects of incineration on the leaching efficiency
[26]	Article	2018	Hydrometallurgy extraction from retired LIBs
[38]	Review	2018	The current status of hydrometallurgy recycling process; pretreatment methods
[115]	Article	2015	Efficient and product-oriented hydrometallurgical recycling of retired automotive batteries
[52]	Review	2021	Overview on the available hydrometallurgical technologies
[113]	Article	2020	Recycling process of LFP-type of LIBs
[116]	Article	2019	LCA; process-based cost model; recycling processes of LIBs
[117]	Review	2017	Current-state of retired $LiFePO_4$ batteries recycling in China
[118]	Article	2014	Physical and chemical treatments for LIBs modules used in hybrid EV
[67]	Article	2001	A laboratory process of LIBs recycling
[34]	Review	2019	Summary of recycling processes of EOL batteries
[119]	Proceeding Paper	2015	Recycling process of retired automotive LIBs
[20]	Proceeding Paper	2020	Optimizing efficient hydrometallurgical processes of LIBs
[120]	Review	2020	Closed-loop strategy for cycling cathode materials; utilization of exhausted anode materials
[57]	Review	2020	Recovery process and products of waste LIBs
[121]	Article	2014	Solid−liquid equilibrium (SLE) phase behaviour and process optimize of retired LIBs
[56]	Review	2021	Processing of organic binders; recycling technologies
[38]	Review	2018	The current status of hydrometallurgical recycling technologies
[36]	Article	2015	Recycling concepts for retired LIBs; state-of-the-art schemes of waste treatment technology

Table A5. Assessed literature in review for "Direct Recycling Process" category.

			Direct Recycling Process
[58]	Review	2020	Retired LIBs recycling processes
[19]	Article	2021	Direct cathode recycling of EOL LIBs
[73]	Article	2020	Industrial model for direct recycling of LIBs
[122]	Article	2020	Removal of the PVDF binder and carbon black through thermal processing
[123]	Article	2019	Characterization of aged components; direct recycling
[124]	Review	2016	The current status of graphite anodes in the present recycling technologies of retired LIBs

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Table A5. *Cont.*

			Direct Recycling Process
[125]	Article	2019	Direct recycling or two-step carbonization for LIBs anode materials
[126]	Article	2020	Revitalization of composition, structure, and electrochemical performance of LFP LIBs with different degradation conditions; LCA
[74]	Article	2021	Mathematical regression model; retrieval efficiency using Taguchi Design of Experiment (DoE) method
[127]	Article	2018	Aged cathode materials; two direct recycling methods: solid-state, and hydrothermal.
[128]	Article	2018	Four recycling steams; evolution of the precursor particles
[129]	Article	2013	Recycling mixed cathode materials
[130]	Review	2018	The whole recycling process; hydrometallurgy
[8]	Review	2020	A systematic overview of rechargeable battery sustainability
[131]	Review	2020	Current recycling status for LIBs; advancements in these methods
[5]	Article	2017	A novel approach to recycling mixed cathode materials based on a closed-loop
[132]	Article	2021	Study of 44 commercial recyclers; a novel qualitative assessment matrix termed "Strategic materials Weighting And Value Evaluation" (SWAVE)

Table A6. Assessed literature in review for "Special Method" category.

			Special Recycling Methods
[81]	Article	2020	Process models of state-of-the-art pyrometallurgical and hydrometallurgical recycling based on real data
[83]	Article	2020	Life-cycle burdens of LIBs
[84]	Article	2011	LCA; benefits of EOF batteries
[82]	Article	2018	Staklberg game theory-based model; reward-penalty mechanisms and policies—a case of Beijing
[85]	Review	2020	Safe recycling, physical processes
[87]	Processing Paper	2016	LithoRec project
[88]	Article	2020	Synthesisation and extension design for recycling (DfR) principles
[92]	Review	2020	Recycling–Molten salt approach
[86]	Article	2018	Recycling processes, dis/advantages
[93]	Article	2017	Mechanochemical process using polyvinyl chloride
[133]	Article	2018	Metal recycling using ammonium chloride
[94]	Article	2020	Metal Organic Frameworks recycling process
[95]	Article	2020	Microwave processing route
[89]	Article	2013	Wet and dry crushing methods
[134]	Article	2021	Recycling using catalytic pyrolysis or gasification of biomass
[91]	Article	2018	Heat-treatment recycling of waste toner
[135]	Editorial Material	2018	Electrohydraulic crushing
[36]	Article	2012	ACCUREC Recycling and UVR-FIA a recycling process specially dedicated to portable LIBs

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Pre-Recycling Material Analysis of NMC Lithium-Ion Battery Cells from Electric Vehicles

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Abstract: Environmental concerns push for a reduction in greenhouse gas emissions and technologies with a low carbon footprint. In the transportation sector, this drives the transition toward electric vehicles (EVs), which are nowadays mainly based on lithium-ion batteries (LIBs). As the number of produced EVs is rapidly growing, a large amount of waste batteries is expected in the future. Recycling seems to be one of the most promising end-of-life (EOL) methods; it reduces raw material consumption in battery production and the environmental burden. Thus, this work introduces a comprehensive pre-recycling material characterization of waste nickel-manganese-cobalt (NMC) LIB cells from a fully electric battery electric vehicle (BEV), which represents a basis for cost-effective and environmentally friendly recycling focusing on the efficiency of the implemented technique. The composition of the NCM 622 battery cell was determined; it included a $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ spinel on a 15 µm Al-based current collector (cathode), a graphite layer on 60 µm copper foil (anode), 25 µm PE/PVDF polymer separator, and a $LiPF_6$ salt electrolyte with a 1:3 ratio in primary solvents DMC and DEC. The performed research was based on a series of X-ray, infrared (IR) measurements, gas chromatography–mass spectrometry (GC-MS), and inductively coupled plasma–optical emission spectrometry (ICP-OES) characterization of an aqueous solution with dissolved electrolytes. These results will be used in subsequent works devoted to optimizing the most suitable recycling technique considering the environmental and economic perspectives.

Keywords: lithium-ion battery (LIB); electric vehicle (EV); recycling process; waste battery; end-of-life treatment (EOL); material analysis

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1. Introduction

Nowadays, lithium-ion batteries (LIBs) represent a crucial energy storage technology [1,2]. They have found their place in various applications, from consumer portable electronics to complex stationary systems for energy storage, including smart grids and microgrids, particularly in the automotive industry [2–9]. LIBs are considered the most efficient technology for large-scale electrical power systems due to their high voltage and energy density, negligible memory effect, fast charging speed, and long cycle life [10,11]. Thus, they represent the heart of energy storage in present-day electric vehicles (EVs) [12,13].

More and more LIB applications are utilizing pouch cell design, an excellent solution for weight and cost reduction and optimizing packaging efficiency at the battery cell level [14,15]. Conductive foils, covered with active electrode materials, are divided with a separator and soaked with electrolytes. Several dozens of these cathode–separator–anode layers together represent one general battery cell, whose material composition is illustrated in Figure 1. The foil tabs are welded to the electrode and sealed to the aluminum (Al) pouch cover, carrying the positive and negative terminals [14,16–19].

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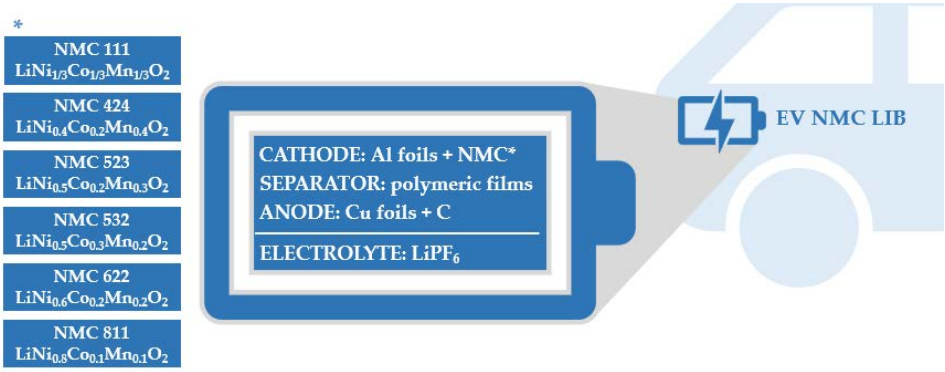

Figure 1. Composition of a general NMC LIB cell (* it complements individual NMC cell chemistries).

Lithium-nickel-manganese-cobalt-oxide ($LiNi_xMn_yCo_zO_2$) cathode material for LIBs, abbreviated as NMC, is today considered one of the most successful technologies [17,19,20]. It benefits from the properties of the blend of nickel (Ni), manganese (Mn), and cobalt (Co), where each metallic element plays a crucial role. The Ni furnishes the efficient capacity, the Co contributes to a good cycling life, and the Mn causes solid structural stability [20]. The NMC blend benefits of several varieties described according to the proportion of transition metal atoms. The basic formula of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ consists of 33% Ni, 33% Mn, and 33% Co, typically referred to as 1-1-1 (NMC 111). The other typically used blends include $LiNi_{0.4}Co_{0.2}Mn_{0.4}O_2$ (NCM 424), $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ (NCM 523), $LiNi_{0.5}Co_{0.3}Mn_{0.2}O_2$ (NCM 532), $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ (NCM 622), and $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ (NCM 811) [17,20–22]. Aluminum (Al) has been widely used as a current collector for LIBs in recent years. It accounts for over 90% of the electric conductivity and approximately 90% of the cathode's mechanical strength in LIB cells [19,23].

The anode of the LIB cell is commonly created by graphite deposited on copper (Cu) foil. Graphite (C) is mainly used due to its reversible ability to position lithium (Li) ions between material layers of the cell. Similar properties are shown by other materials, such as activated carbon, carbon black, conductive additives, lithium titanate (LTO), silicon, or graphene [18,19,24].

Polyolefine (PO) membrane technology usually serves the LIB cell separator [25]. Depending on the manufacturing process, either polyethylene (PE) polymer and wax ("wet process") or PE or/and polypropylene (PP) polymer ("dry process") is used. These polymeric organic thin films can be additionally treated; for example, the polyvinylidene difluoride (PVDF) particles are used for obtaining strong polarity, stable electrochemical performance, and high thermal stability [25,26].

Nowadays, LIB manufacturers are focused on liquid electrolytes. A very common NMC LIB electrolyte is a 1-molar solution of highly conductive lithium hexafluorophosphate salt ($LiPF_6$) dissolved in various ratios of Ethylene Carbonate (EC):Diethyl Carbonate (DEC) and 2% Vinylene carbonate (VC) [27]. Although this electrolyte has excellent conductive properties, it dissociates into lithium fluoride (LiF) and phosphorus pentafluoride (PF_5), which reacts with moisture and forms hydrogen fluoride (HF) and phosphoryl fluoride (OPF_3) [25,28]. HF can lead to electrode material reactions that lose the power and capacity of the battery cell and represent high environmental and health risks due to the high acute toxicity, e.g., during end-of-life (EOL) processing [27,28].

The global targets of greenhouse gas emission reduction are rapidly driving the demand for alternative fuel vehicles with the benefit of a low carbon footprint [29–33]. Nevertheless, the still-increasing numbers of recently produced EVs bring new issues that must be solved; one of the most critical topics is the EOL processing of the ever-increasing

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number of waste batteries [34,35]. Currently, the most prospective procedure is the recycling of waste batteries and the recovery of valuable and essential metals, such as Ni, Mn, Co, Li, Co, and Al [34,36–39].

Pyrometallurgy and hydrometallurgy are highly implemented techniques in recycling. The pyrometallurgical method is based on thermally treated batteries' physical and chemical transformation, with the process efficiency between 80–98.9% and 98–99.95% output purity. The hydrometallurgical process is created by chemical treatment, including acid or basic leaching, with an efficient process of around 76–98.2% and high-purity products between 96.5–99.7% [34,40–43]. The process efficiency entirely depends on the investment costs and equipment of the recycling plant [33,40]. Thus, the EU has responded to this critical automotive industry change and introduced new emission-free standards and regulations considering the real waste treatment solutions, determining the recycling efficiencies for LIBs to reach 65% by 2025 and 70% by 2030 [44].

Since there are many different EV LIBs on the market, it is necessary to understand the selected technology in its waste state before integrating the recycling process. Thus, this work contains a detailed pre-recycling material analysis of waste nickel-manganese-cobalt (NMC) LIB cells from a fully electric battery electric vehicle (BEV), which represents a basis for the effective recycling to meet the high EU requirements. Based on the performed X-ray and infrared (IR) spectra measurements, the cathode (Al foil and $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ spinel), anode (Cu foil and graphite layer), and separator (PE/PVDF thin film) materials were determined. Gas chromatography–mass spectrometry (GC-MS) and inductively coupled plasma–optical emission spectrometry (ICP-OES) were used for complete electrolyte characterization from the aqueous solution. The $LiPF_6$ salt with the main ratio of solvents 1:3 DMC:DEC was established for the cell. Moreover, the battery technology's valuable metals blend was set the closest to a $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ (NCM 622).

The content of this work is structured as follows: in Section 2, the detailed overview of used materials and methods is provided; in Section 3, the obtained results of NMC LIB cell material characterization are presented; in Section 4, the contribution of this work and its context of LIBs recycling in terms of future work is discussed; in Section 5, the main conclusions of this paper are pointed out.

2. Materials and Methods

The degradation process was determined using material analysis characterization on the waste nickel-manganese-cobalt (NMC) cells dismantled from a fully electric battery electric vehicle (BEV) module.

2.1. Materials

The waste NMC LIB pouch cells were used for laboratory testing, with each cell having a nominal voltage of 3.65 V and 78 Ah capacity. These cells were dismantled from the short-circuited battery module, commonly used in EV battery packs. The module consists of 24 cells in a 3P8S connection and a nominal voltage of 29.36 V and 6.85 kWh capacity.

2.2. Methods

This work used experimental methods for material analysis, such as X-ray diffraction (XRD) and fluorescence (XRF), infrared spectroscopy (IRS), light and electron microscopy, including mode of secondary electron (SE), and energy dispersive spectrometry (EDS). Moreover, one cell was opened and fully disassembled in 12 L of tap water to reduce the potential safety hazards associated with $LiFP_6$. The obtained aqueous solution was characterized in detail for determining the composition of the electrolyte using gas chromatography–mass spectrometry (GC-MS) and inductively coupled plasma–optical emission spectrometry (ICP-OES).

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2.2.1. Material Analysis

This work is based on our previous work [45]. Moreover, a detailed analysis of electrodes, separator, and electrolyte from the light and electron microscopy, infrared spectroscopy (IRS), and spectrometry field was added. The performed methods used for the individual battery cell components' material characterizations are shown in the diagram according to Figure 2.

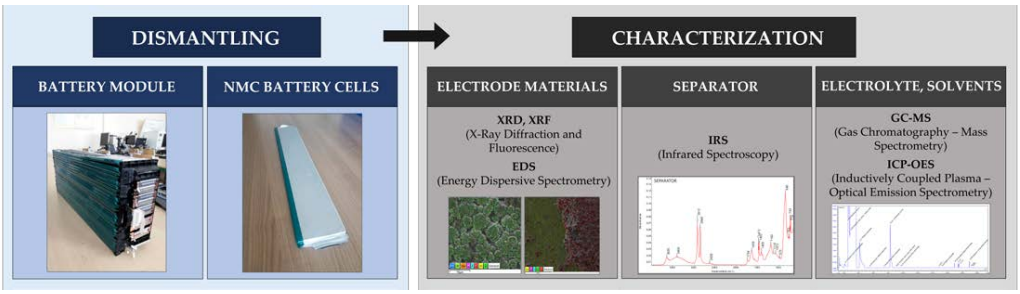

Figure 2. Material analysis methods used for battery cell characterization.

X-ray Diffraction and Fluorescence

The material of thin layers on metal sheets or a cell separator were analyzed using an X-ray fluorescence (XRF) with a Philips PANAlytical spectrometer and X-ray diffraction (XRD) with a Philips PANAlytical XPERT-PRO diffractometer.

Light and Electron Microscopy

Analyses of the thin films were carried out using an Olympus LEXT OLS5000 SAF confocal microscope at 5×, 10×, 20×, 50×, and 100× magnifications. A laser surface scanning mode was used for detailed illustration. Layer width measurements were performed in NIST Elements software.

Furthermore, the structures were characterized with a scanning electron microscope SEM TESCAN VEGA 3 LMU at an accelerating voltage of 10 kV and a working distance of 15 mm. It disposes of a ±0.1% error for each point of the relative error for a performed measurement. Surface morphologies were recorded in a mode of secondary electron (SE) and backscattered electrons (BSE) at a magnification of 300×, 500×, 2000×, and 3000×. In addition, a micro-X-ray elemental map was created using energy dispersive spectrometry (EDS).

Infrared Spectroscopy

The infrared (IR) ATR FTIR spectra were analyzed using a Thermo Scientific Nicolet 6700 spectrometer using a PIKE Technologies GladiATR reflective ATR extension with a diamond crystal. Spectra were recorded in the 4000–400 cm^{-1} region with a DLaTGS (deuterated L-alanine doped triglycine sulfate) detector with 4 cm^{-1} resolution, 64 scans, and Happ-Genzel apodization.

Gas Chromatography—Mass Spectrometry

Volatile solvents were characterized by a Thermo Fisher Scientific Trace 1310 GC coupled to a ISQ 7000 mass spectrometer. An autosampler TriPlus RSH used an SPME arrow (acrylate) extraction for the comprehensive identification of organic compounds present in the sample, and to estimate carbonate ratio headspace injection was performed. Both sample injection techniques preceded the incubation of a 1 mL sample at 80 °C for 30 min. GC separation ran on semi-standard non-polar column TG-5SILMS (30 m; 0.25 mm; 0.25 µm) with helium as a mobile phase (1.2 mL/min). The oven program started at 40 °C

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held for 1 min followed by a 5 °C/min gradient up to 280 °C. A single quadrupole mass spectrometer was set to full scan acquisition with 5 Hz scan rate.

Inductively Coupled Plasma—Optical Emission Spectrometry

For quantitation of each element in aqueous solution containing electrolyte, Perkin Elmer Optima 8000 was set to monitor emission bands of Li (670.784 nm), Ni (231.604 nm), Mn (257.610 nm), Co (220.616 nm), Cu (327.393 nm), Al (396.153 nm), and P (213.617 nm). The samples were centrifuged and diluted 10× into 0.1% HNO_3, 100× if out of linear range.

3. Results

Pouch cells are made up of alternately layered cathode and anode materials, which are split by separators. Thus, firstly in this work, the cathode (on Al foil), anode (on Cu foil), and separator samples were analyzed. The tested NMC pouch cell consists of 36 spinel-separator-graphite cells. The analyzed layer composition with the determined materials' thicknesses is illustrated in the structure scheme in Figure 3. Within these characterization measurements, one side of each metal foil was covered by a visually pure material layer, and on the second side a layer was deposited containing remains of the separator, which were confirmed by energy dispersive spectroscopy.

Figure 3. Scheme of the analyzed NMC battery cell structure.

The XRD and XRF methods supplemented with EDS element maps were used for the determination of the stoichiometric composition of the materials; the IR was performed for the separator analysis. ICP-OES, and GC-MS were performed for the analysis of the dissolved electrolyte in an aqueous solution. The results, including average thickness and experimental method for the individual material, are shown in Table 1.

The cathode active material layer is thick, approximately 175 µm; it is formed by a 15 um Al foil and an 80 µm spinel layer adjoined on each side. A detail of the material thickness is shown in Figure 4a.

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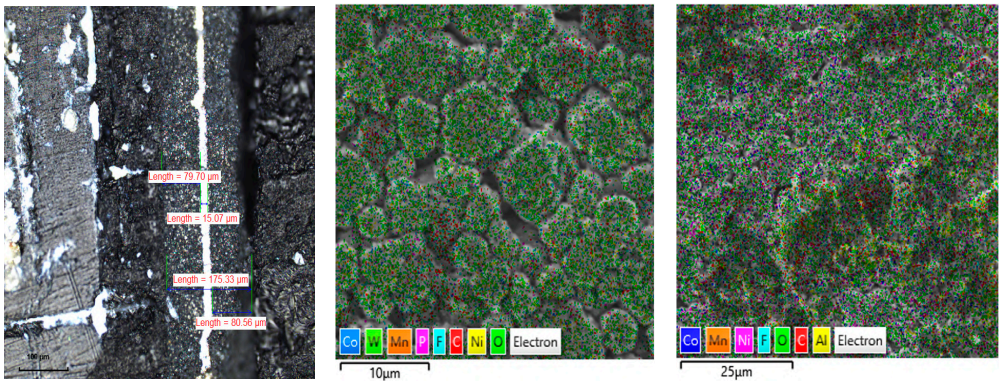
Table 1. The material composition of the individual components of the battery cell; average thicknesses of individual material layers; methods used to characterize the material composition.

Battery Cell Component	Material Composition	Material Thickness	Analysis Method
Cathode	Al foil $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$	15 µm 80 µm in one layer	XRD, XRF, EDS
Anode	Cu foil carbon	60 µm 100 µm in one layer	XRD, XRF, EDS
Separator	PE/PVDF	25 µm	IR
Electrolyte	$LiPF_6$	-	ICP-OES
Solvents	Carbonates, FB	-	GC-MS



(**a**) (**b**) (**c**)

Figure 4. The cathode of the battery cell: (**a**) cross-section of a spinel double layer on Al foil with an illustration of materials layer thickness; (**b**) EDS element map of visually pure side of spinel layer; (**c**) EDS element map of spinel layer with separator residues.

The surface of the cathode material samples was X-rayed in EDS; the elemental maps for the visually pure spinel layer and the layer with separator residues are shown in Figure 4b,c. Based on the results shown in Table 2 and the XRF measurement, with the ratio between Ni:Mn:Co in 55:24:20 mol.%, the NMC 622 blend was determined for the spherical spinel particles of the tested battery cell. The presence of tungsten (W) was confirmed by both EDS and XRF analysis. It is probably an additive in the form of, e.g., tungsten trioxide (WO_3), to improve the cathode materials' electron conductivity and redox reaction kinetics as illustrated by Meng et al. [46] or Gan et al. [47].

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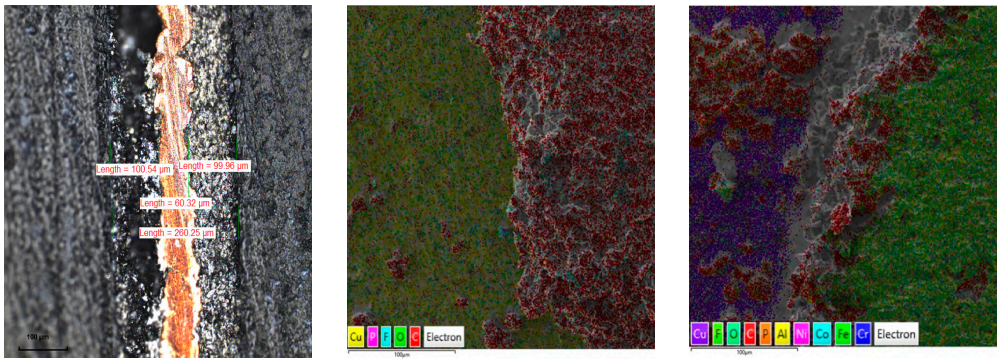
Table 2. Results of the EDS element map for the battery cathode.

Element	Visually Pure Layer		Layer with Separator Residues	
	Weight (wt.%)	σ (wt.%)	Weight (wt.%)	σ (wt.%)
Ni	28.0	2.7	23.1	2.2
O	23.7	1.1	21.7	0.8
F	16.4	0.8	16.7	0.7
C	10.9	0.7	15.0	0.7
Mn	8.5	0.9	7.4	0.7
Co	7.3	1.6	7.8	1.3
W	3.8	0.5	1.5	0.3
P	1.3	0.2	0.6	0.1
Al	-		6.1	0.3

The entire carbon bilayer deposited on a Cu (anode) is around 100 µm thicker than the layer of cathode materials (it measures 260 µm in total). The Cu foil has an average of 60 µm, with each spinel layer 100 µm thick. A detail of the active material thickness is shown in Figure 5a.



(**a**) (**b**) (**c**)

Figure 5. The anode of the battery cell: (**a**) cross-section of a graphite double layer on Cu foil with an illustration of materials layer thickness; (**b**) EDS element map of visually pure layer; (**c**) EDS element map of graphite layer with separator residues.

The sample surface of anode materials was X-rayed in EDS mode; the elemental maps for the visually pure layer and spinel with separator residues are shown in Figure 5b,c, respectively. The elemental analysis shown in Table 3 detected electrolyte residues (containing F and P) and confirmed the presence of a separator (based on Al). The material layer was determined to be >90% pure graphite based on the results of XRF analysis.

Table 3. Results of the EDS element map for the battery anode.

Element	Visually Pure Layer		Layer with Separator Residues	
	Weight (wt.%)	σ (wt.%)	Weight (wt.%)	σ (wt.%)
C	48.0	0.6	35.8	0.7
Cu	39.1	0.6	23.0	0.4
O	6.0	0.2	14.1	0.3
F	4.3	0.2	10.4	0.2
P	1.1	0.1	2.6	0.1
Al	-		12.9	0.1

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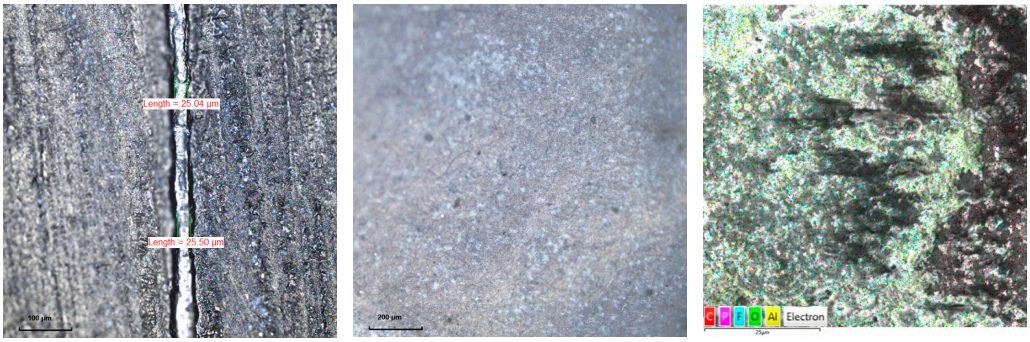
The battery separator macroscopically appeared as a rigid, flexible polymer thin film of an approximate thickness of 25 µm. A detail of the separator cross-section and an optical image of the structure are shown in Figure 6a and b, respectively.

Due to the separator samples' high fluorescence, it was impossible to use Raman spectroscopy to determine their composition. Therefore, the separator's IR spectrum was measured, as shown in Figure 7.



(**a**) (**b**) (**c**)

Figure 6. The separator of the battery cell: (**a**) cross-section with an illustration of material layer thickness; (**b**) optical image from a confocal microscope; (**c**) EDS element map.

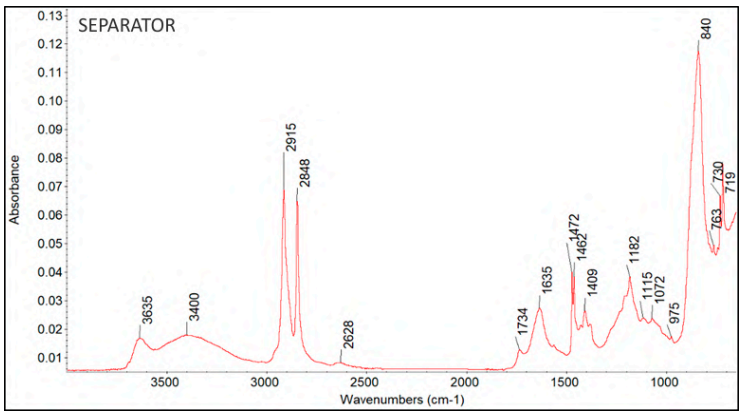


Figure 7. IR spectrum of the separator.

To interpret the spectrum, polyethylene film (PE) was used as a reference material and was measured under the same conditions (Figure 8). Comparing these two spectra concluded that the absorption bands at 2915, 2848, 1472, 1462, 730, and 719 cm^{-1} correspond to the polyethylene polymer.

To interpret the whole spectrum, individual absorption bands were identified using reference materials, spectrum libraries, and the available literature. The main component of the separator material was determined as the PE/PVDF polymer. The absorption bands at 1409, 1182, 1072, and 840 cm^{-1} correspond to the polyvinylidene fluoride. The other absorption bands were assigned to hexafluorophosphate $[PF_6]^-$, aluminum oxide (Al_2O_3), and residual carbon. The elements of all materials were confirmed by elemental analysis,

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and the identified materials correspond with previously published results, where the PE separator with surface coating modification is commonly used for LIBs due to its high thermal dimensional stability and excellent electrolyte wettability. The properties of the same type of separator (PE separator coated by PVDF particles) were discussed in work by Wang et al. [48].

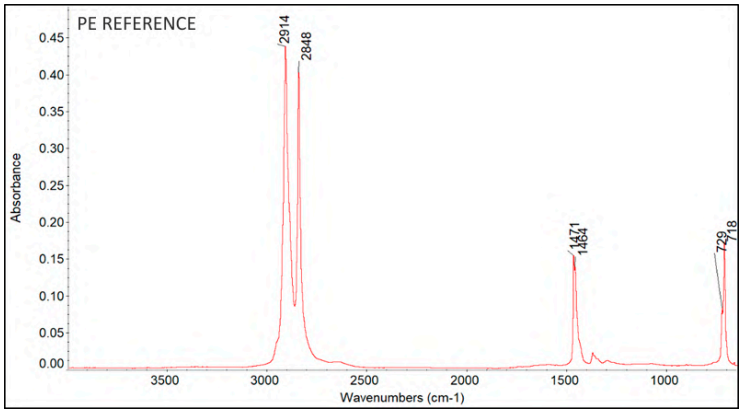

Figure 8. IR spectrum of the polyethylene reference foil PE Reference.

These results support the presence of C, Al, and F atoms detected by the EDS element map, of which the results are shown in Figure 6c and in Table 4.

Table 4. Results of the EDS element map for the battery separator.

Element	Separator	
	Weight (wt.%)	σ (wt.%)
F	31.9	0.5
C	29.2	0.7
O	19.8	0.4
Al	15.7	0.3
P	3.5	0.2

Based on a performed characterization, lithium hexafluorophosphate ($LiPF_6$) electrolyte residues, containing F and P, were determined on both electrodes using EDS spectrometry mode and on a battery separator by IR spectra measurement.

Regardless, the electrolyte composition was analyzed in the second part of this work. The electrolyte solvents from the aqueous solution were analyzed in detail using GC-MS and ICP-OES. The cell was cut open in water, disassembled from the Al package, and hand-crushed, and then the coarse solid parts were removed from the solution. The results of the analysis are shown in Table 5.

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Table 5. Electrolyte and electrode dissolution in aqueous solution.

Concentration (mg/L)	Al	Co	Cu	Li	Mn	Ni	P
Blank	<LOQ	<LOQ	1.6	0.1	<LOQ	<LOQ	1.7
Cell opening (time: 0:00)	<LOQ	<LOQ	2.0	12.5	<LOQ	<LOQ	764.8
Cell disassembly (time: 0:30)	2.5	0.1	18.8	110.5	0.3	0.4	4 316.8
Cell crushing (time: 0:45)	3.3	0.1	18.0	106.4	0.4	0.5	4 139.8
Removal of solids (time: 1:00)	8.6	0.2	17.6	133.1	0.8	1.1	4 501.1
mg/one cell	103.0	3.0	210.8	1 597.6	9.5	13.8	54 012.8

LOQ = limit of quantification.

A trace amount of Co, Mn, and Ni was dissolved, whereas Al and Cu were released from the solid phase in a slightly higher amount. The most abundant element observed was phosphorus (P), most likely phosphates. The amount of Li observed is significantly lower than P, probably due to the formation of insoluble lithium salts.

The most abundant solvent identified according to the chromatogram illustrated in Figure 9 was methyl ethyl carbonate (EMC), followed by its dimethyl and diethyl esters; a significant amount of fluorobenzene (C_6H_5F) was observed.

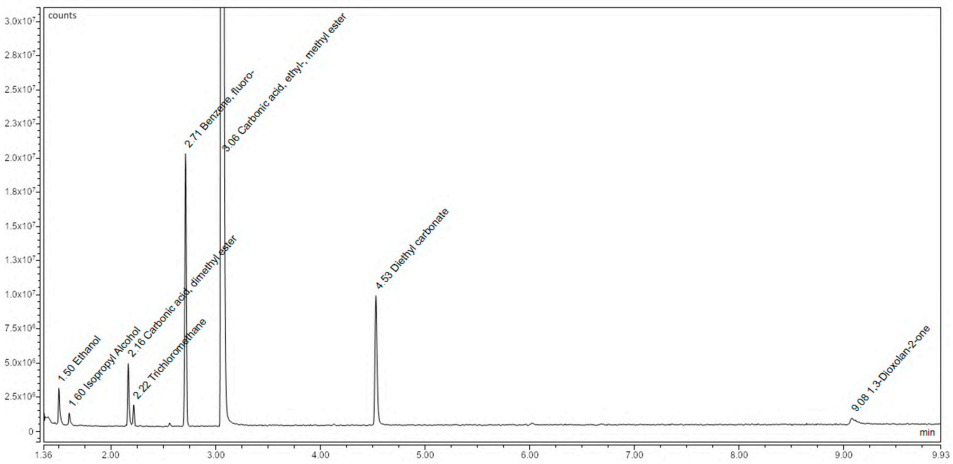

Figure 9. Chromatogram of headspace injection for estimation of carbonate ratio and identification of major organic solvents.

Solid-phase microextraction (SPME) revealed trace amounts of other carbonic acid esters with various carbon chain lengths, as visible in Figure 10. All these compounds have high vapor pressure and migrate into the air.

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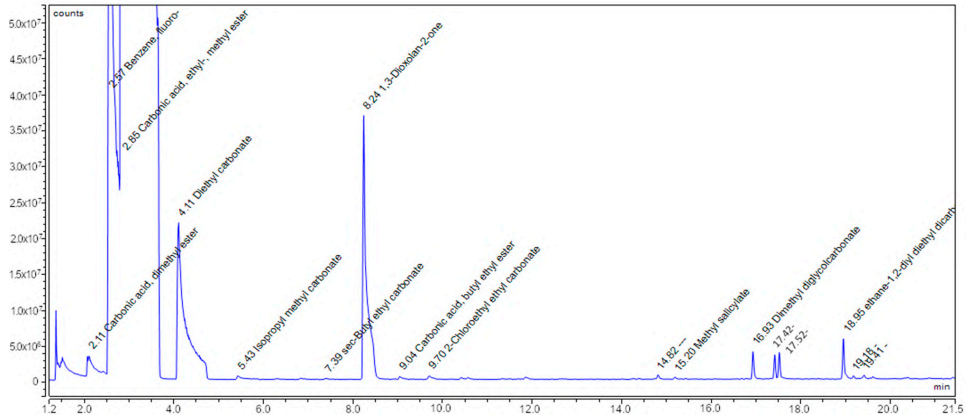

Figure 10. SPME extraction of volatile solvents from aqueous solution, further identification of trace carbonates.

The ratio for the primarily used solvents for $LiPF_6$ salt electrolyte, dimethyl carbonate (DMC), and diethyl carbonate (DEC), was established as 1:3. These results were determined for the waste NMC cell based on the electrolyte release into an aqueous solution.

Based on the experimental results of this work, LIB NMC pouch cells consisting of 36 spinel-separator-graphite cells in total were characterized in detail. According to the performed analyses, including XRD, XRF, EDS, and IR measurements, the cathode of the battery cell is created by the 15 µm Al-based current collector, which is covered from both sides by the 80 µm NMC 622 spinel. The battery anode is made up of 60 µm Cu foil, with graphite layers deposited on both sides with a >90% purity. The cathode and anode materials are split by a separator based on PE/PVDF polymers, ensuring high thermal dimensional stability and excellent electrolyte wettability. The battery cell electrolyte was determined based on a $LiPF_6$ salt with a 1:3 ratio in primary solvents DMC and DEC with MEC being the main component; the characterization was set up by GC-MS and ICP-OES measurements of an aqueous solution containing released electrolyte and active material residues.

4. Discussion

This work represents an important step in the EOL processing of NMC LIB technology, as it characterizes in detail the material composition of the waste cell. During operation, the active materials of the electrodes, current collectors, separator, and electrolyte degrade, i.e., change their capabilities. Therefore, it is necessary to characterize the state of cells at the beginning and end of their life to precisely define the input products of recycling processing.

The results of this work create a basis for future work, as shown in Figure 11. Detailed material characterization of the NMC technology will be used to optimize the individual steps of currently used recycling techniques, focusing on achieving the highest possible process efficiency and purity of recovered materials whilst considering the investment and operating costs. Moreover, the environmental impact based on the performed analysis results will be described, including wastewater treatment (containing toxic electrolyte residues) or emissions generated during high-temperature processing. The work will also focus on recovery from secondary processes, e.g., wastewater, which would directly support the principle of lossless recycling.

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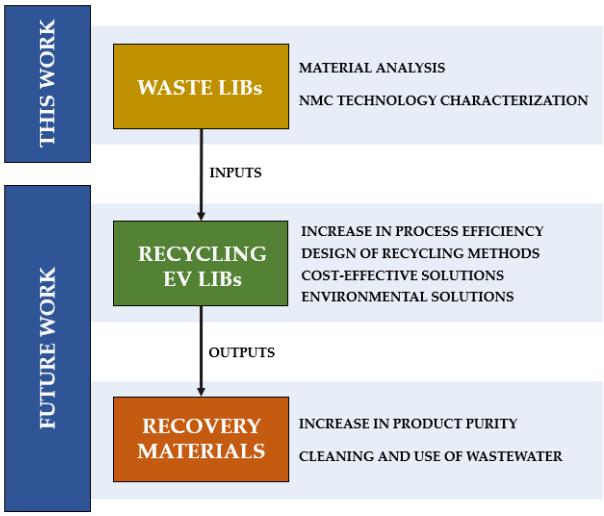

Figure 11. Inclusion of this work in the context of LIBs recycling and follow-up work.

The performed analysis was carried out to characterize waste battery cells' material state before recycling. Thus, fully discharged (short-circuited) cells were used for evaluation; this could have degraded and changed the structure of the materials. Thus, the obtained material analysis might not be completely valid for a fresh cell.

Although the market for LIBs is still growing, understanding the currently widely used technologies is a necessary basis that will lead to the successful compliance with the high EU requirements for EOL processing and faster adaptation when processing new technologies.

5. Conclusions

This work performed a detailed pre-recycling material analysis of the LIB NMC cell, commonly used in a BEV. Results of the presented battery cell material composition can be used as a basis for better understanding the degradation mechanism of NMC technology, process modeling, and for implementing ecologically and economically beneficial EOL processing, including high-efficiency recycling.

Detailed characterization results of LIB NMC cells, including analysis of the active electrode materials, separator, and electrolyte composition, were presented in this paper. The results will be followed up in subsequent works. Determining the exact material ratios of the waste battery pouch cells, established as NMC 622 (based on $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ spinel), will be used to elucidate the cell's degradation mechanisms, focusing on electrode material changes and electrolyte decomposition, compared with analyses of the new cell technology being tested.

Moreover, this work represents a necessary step that precedes recycling the entire battery cell. The results will be used in the follow-up work, devoted to selecting and optimizing the most suitable recycling technique focusing on recovering the highest possible proportion of crucial materials and their sufficient quality when reused in EV LIB cell production. Moreover, in terms of recycling, the explicit materials characterization will be part of the process modeling, considering the economic and environmental impacts of implementing recovery techniques.

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Author Contributions: Conceptualization, A.P. and J.K.; methodology, A.P.; formal analysis, J.K.; investigation, J.K. and M.H.M.; resources, A.P., J.K., M.H.M., Z.P. and D.P.; data curation, A.P. and J.K.; writing—original draft preparation, A.P.; writing—review and editing, A.P., M.H.M., J.K., D.P. and V.K.; supervision, M.H.M. and V.K.; project administration, A.P. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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Thermal stability of valuable metals in lithium-ion battery cathode materials: Temperature range 100–400 °C

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HIGHLIGHTS

- First step of pyro-hydrometallurgical process: heat treated cathode at 100–400 °C.
- ICP-OES analysis of valuable metals (Li, Ni, Mn, Co).
- Analysis of NMC spinel using XRF, XRD, TGA, SEM/EDS.
- Leaching efficiency of NMC in demineralized water and nitric acid.
- Comparison of yield with other recycling techniques.

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ABSTRACT

Lithium is crucial in lithium-ion batteries (LIBs), serving as a main component of the electrolyte and cathode. Elements such as cobalt, nickel, and manganese are also vital for high performance, energy density, and stability. This study aimed to examine the behaviour of end-of-life cathode material ($LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$) and its valuable metals after exposure to temperatures between 100 and 400 °C, comparing it with untreated material. The lithium content cannot be reliably determined by conventional analytical methods, so inductively coupled plasma optical emission spectroscopy (ICP-OES) was chosen for this purpose. For ICP-OES measurements, samples were dissolved in different solvents for a specified time, and the concentrations of lithium, nickel, manganese, and cobalt were measured. From the measured values, their theoretical yields were calculated. Due to the annealing at given temperatures and subsequent dissolution, this step can be considered as the first stage of the pyrometallurgical-hydrometallurgical process used in battery recycling. The study was complemented by further analyses to monitor the effect of annealing temperatures on the properties of the material. Based on the results, it was found that the highest theoretical yield in this temperature range was for material annealed at 400 °C and dissolved in 20 % nitric acid for 4 h.

1. Introduction

Lithium-ion batteries (LIBs) are considered the most powerful energy storage system for portable electronic devices, wireless devices, hybrid power and electric vehicles (EVs) due to their long life, high energy density, and high voltage [1–3]. For this reason, the LIB industry is one of the fastest growing industries in the world, but the scarce raw materials such as lithium, nickel, manganese, and cobalt make it essential to manage these resources effectively. There are various approaches to sustainability in this sector, including battery reuse and recycling. As the amount of spent batteries increases, the waste management will need to be systematically addressed. Recycling is becoming crucial for the renewability of scarce elements [4–6]. An efficient way to dispose of spent LIBs is a key factor for industrial battery manufacturers

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and recyclers. This is important due to the limited resources of strategic metals, the potential negative environmental impact, and the potential to obtain high economic benefits [2,6].

The basic description of a battery cell involves the use of the relationship between the cell potential and the concentrations of individual components. This is described by the Nernst equation (see Equation (1)),

$$\Delta E = \Delta E^0 + \frac{kT}{v_e e} ln \frac{a_{Ox}}{a_{Red}} \quad (1)$$

where k [JK^{-1}] represents the Boltzmann constant, T is the temperature [K], v_e is the number of electrons exchanged, and e stands for elementary charge [C]. The electrode potential ΔE is then described using the natural logarithm of the ratio of chemical activities of the relevant oxidised and reduced components in the system and the standard electrode potential ΔE^0 [7,8].

In essence, LIBs can be defined as energy storage systems that operate through the process of lithium-ion insertion reactions occurring at both the positive and negative electrodes, where lithium ions serve as the charge carriers. This broad categorisation encompasses various cell chemistries that collectively constitute the LIB family. The majority of LIBs typically feature a negative electrode primarily composed of materials such as carbon (e.g., graphite), lithium titanate ($Li_4Ti_5O_{12}$) or innovative materials like metallic lithium and Li(Si) alloys. The choice of electrolyte can vary depending on the electrode materials selected, but typically comprises a blend of lithium salts (e.g. $LiPF_6$) and an organic solvent (e.g. diethyl carbonate) to facilitate ion transfer. To facilitate the passage of lithium ions between electrodes while preventing the occurrence of internal short circuits, a separating membrane is employed [9–11]. Known examples of materials used as cathodes include lithium cobalt oxide ($LiCoO_2$), lithium iron phosphate ($LiFePO_4$) (which can be coated with Ga or SiO_2), lithium nickel manganese cobalt oxide ($Li(Ni_{1-x-y}Mn_xCo_y)O_2$), lithium nickel cobalt aluminium oxide ($Li(Ni_{1-x-y}Co_xAl_y)O_2$), and lithium manganese oxide ($LiMn_2O_4$) [12–15].

The predominant cathode materials used in LIBs for EVs are part of a category of layered mixed transition metal oxide compounds characterised by rhombohedral symmetry (D^5_{3d} space group). These compounds are represented by the formula $LiNi_xMn_yCo_{1-x-y}O_2$ (with $(x + y) \epsilon [0,1]$) and are commonly called NMC [16–18]. The principle of NMC cathode materials lies in the combination of nickel and manganese, each contributing specific attributes. Nickel is known for its high specific energy, but has the drawback of poor stability, while manganese possesses the advantage of forming a spinel structure to reduce internal resistance, though it offers lower specific energy. The specific combination of these metals, nickel and manganese, varies among manufacturers and is closely guarded as a proprietary formula. Researchers are actively exploring nickel-rich electrodes to enhance energy density while reducing the cobalt content to reduce costs. Companies have transitioned from NMC111 to NMC442, NMC622, NMC721 and now anticipate the introduction of NMC811 [9,19,20].

The theoretical capacity of stoichiometric NMC materials is around 275 mAh • g^{-1}, though its accessible capacity depends on the ratio of transition-metal ions within the material. Within the layered transition metal oxide category, cathodes rich in nickel, such as NMC811, exhibit higher capacity and operational voltage, with nickel as the primary active redox species ($Ni^{2+} \leftrightarrow Ni^{4+}$). However, nickel-rich compounds often exhibit poor thermal stability. Manganese plays a crucial role in the preservation of battery cycle life and safety, while cobalt enhances electronic conductivity, improving rate capability. Commercially, NMC111 and NMC333 have reached a more mature stage, incorporating the benefits of various transition metals [21]. For nickel-rich NMC materials, the accessible capacity is significantly higher than NMC111 but faces challenges such as cation mixing [22], surface reconstruction to rock salt [23], phase transformation [24], particle cracking [25], transition metal dissolution [26] and parasitic reactions with electrolyte [27].

Cathode scrap stands out as the most precious element for recovery in the context of spent LIBs, given its abundance of valuable metals such as lithium, nickel, manganese, and cobalt. Consequently, a well-defined framework has been put in place to facilitate its retrieval. Nowadays, the techniques employed to recycle cathode scrap mainly encompass hydrometallurgical methods, pyrometallurgical methods, combined pyrometallurgical-hydrometallurgical methods, and innovative techniques such as molten salt, deep eutectic solvent methods or fluorine-rich hydrogel for extracting Li^+ from Li-containing solutions [1,28–31].

Sieber et al. conducted experiments on identical materials using a wide range of acids, such as HCl and HNO_3, as well as other chemicals like citric acid and NaOH. The samples were leached at laboratory temperature with constant stirring. A more sophisticated method to achieve higher yields involved the use of a high-pressure microwave digestion system. The solutions containing dissolved valuable metals were analysed using ICP-OES. Our proposed method similarly aims to increase process yield but does so by pre-treating the cathode material with thermal stress. Lower temperatures were selected to avoid unnecessary increases in environmental and economic costs. Yang et al. also employed thermal pre-treatment at 650 °C followed by leaching in H_2O_2 at 90 °C, a method we sought to avoid due to its energy intensity. Furthermore, Yang et al. did not consider the efficiency of this process for recovering lithium, which we believe is a critical element for the overall efficiency of the process. Our study emphasizes the significance of lithium recovery and strives to present a more sustainable and economically viable approach by optimizing pre-treatment conditions and leaching processes [32,33].

This work focused on the behaviour of the end-of-life cathode material NMC622 from LIB cell when exposed to different temperatures. The key point was to determine the content of valuable metals that are released from the material with increasing temperature. This can then determine the temperature at which the highest yield of valuable metals will be obtained for possible further processing. Therefore, inductively coupled plasma optical emission spectroscopy (ICP-OES) was chosen as the main analysis, which is one of the few methods that can quantitatively determine the lithium content of the material. Other monitored elements were nickel, manganese, and cobalt. A whole set of samples was prepared and annealed for 1 h each at temperatures of 100–800 °C, however, due to the number of results obtained, this paper focuses only on temperatures of 100–400 °C and the results dealing with the behaviour of the material at temperatures of 500–800 °C will be described in following article. For ICP-OES measurements, the material was dissolved in two different solvents, namely nitric acid and demineralized water. Demineralized water was chosen due to its safety, low economic cost, and also because lithium has the potential to be recovered by water. Material that has not been exposed to heat was used as a reference sample. In order to get a comprehensive picture of the material behaviour at different temperatures, other methods were used, namely X-ray diffraction (XRD) and fluorescence (XRF), thermogravimetric analysis (TGA), and scanning electron microscopy with energy dispersive spectroscopy (SEM/EDS). Thus, the results described below represent a comprehensive material analysis of the end-of-life NMC622.

2. Material and methods

2.1. Materials

2.1.1. Chemicals

Nitric acid (HNO_3) used in this study was of analytical grade and was purchased from Lach-Ner, s.r.o. (Neratovice, Czech Republic). The single element lithium standard (99,99 % Li_2CO_3 in 2 % HNO_3, ANALYTIKA, s.r.o., Czech Republic) and the multi-element standard of 26 elements (in 5 % HNO_3, ANALYTIKA, s.r.o., Czech Republic) were used as calibration standards for ICP-OES measurements.

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2.2. Methods

2.2.1. Disassembly procedure

The LIB module, which is commonly used in battery electric vehicles (BEVs), was selected for material analysis. This module consists of 24 NMC pouch cells, each cell has a voltage of 3.65 V and a capacity of 78 Ah, which create the base for the nominal module voltage of 29.36 V. The module was short-circuited to ensure safe handling prior to disassembly, and was then manually disassembled into its individual parts, i.e. milling off the aluminium casing, removing the polyurethane and polymer materials used as electrical and thermal insulation. The remaining plastic and metal coverings were then removed and the individual cells separated [34].

2.2.2. Sample preparation

The LIB pouch cell was disassembled into individual layers of cathodes, anodes, and separators. The cathode material including the aluminium foil was manually cut into square pieces with a size of 25 × 25 mm. These samples were placed in a porcelain crucible and heat treated in an electric furnace at a temperature of 100, 200, 300 and 400 °C for 60 min with a temperature gradient of 5 °C/min in air atmosphere. The samples were labelled according to the firing temperature (e.g. a sample treated at 100 °C for 1 h is named NMC101, etc.). A reference sample that was not exposed to heat treatment is labelled as NMC0. For ICP-OES analysis, the burnt cathode material was finely cut with scissors into strips with a defined area of 17.5 ± 2.5 mm^2. The samples were then dissolved in 25 mL of demineralized water or 20 % nitric acid solution, respectively, for 1.5, 2 and 4 h. The suspension was diluted 500 times, filtered, and analysed by ICP-OES.

All samples were analysed using X-ray diffraction and fluorescence. Samples designated as NMC0 through NMC301 were examined as compact materials due to the difficulty in separating them from the aluminium current collector. In contrast, the sample designated as NMC401 was analysed in powdered form. The NMC401 sample was placed in 80 mL of distilled water in a plastic container. A SONOPULS HD 2200.2 ultrasonic homogeniser with an ultrasonic generator GM 2200.2 and an ultrasonic converter UW 2200 (all BANDELIN, Germany) was used for separation. The amplitude was set to 30 %, the sonication time was 2 × 5 min. The distilled water was poured through a sieve to separate the aluminium parts, and the poured distilled water containing NMC particles was evaporated in an evaporating dish in an oven at 95 °C to constant weight. The dried powdered material was ground in an agate pestle and mortar to obtain a fine powder. The NMC0 sample could not be separated, so it was analysed as a compact sample.

2.2.3. X-ray fluorescence analysis

X-ray fluorescence analysis was conducted using an Axios sequential wavelength-dispersive X-ray fluorescence (WD-XRF) spectrometer. The instrument used for the analysis was equipped with an Rh anode end-window X-ray tube of type 4 GN, featuring a 75 μm Be window. Acquisition of peak intensity data was performed using the SuperQ 5.0 software. The measurements were made under vacuum conditions. The generator settings-collimator-crystal-detector combinations were optimised for collecting 11 scans for all 83 measured elements, for many elements are more than one X-ray line available. The collected data were subsequently processed and quantified using Omnian standard analysis software. The measurement time was about 20 min.

2.2.4. Scanning electron microscopy

The scanning electron microscope TESCAN VEGA 3 LMU (SEM) in secondary electron (SE) mode was used to determine the composition of the materials used and measure their particle size (VEGA3 software). All images were taken at an accelerating voltage of 20 kV, with a working distance of 15 mm and a magnification of 2000×. The OXFORD Instruments INCA 350 EDS analyser enables chemical microanalysis of the observed sample, which is then evaluated in the AZtec software version 4.4. The composition was repeatedly measured on an area of 0.005 mm^2.

2.2.5. X-ray diffraction analysis

X-ray powder diffraction measurements were conducted at room temperature using a PANanalytical X'Pert PRO θ-θ powder diffractometer featuring parafocusing Bragg-Brentano geometry, and CoK_α radiation (λ = 1.79028 Å) at a 35 kV X-ray tube voltage and a current of 40 mA. Data acquisition utilised a high-speed detector X'Celerator. Subsequent data analysis and interpretation were carried out using the software package HighScore Plus 4.0.

2.2.6. Thermal properties

Thermogravimetric measurement was performed on Discovery TGA550 Auto Advanced in air atmosphere (air, technical grade, SIAD s.r.o.) at heating rate 10 °C/min from room temperature to 1000 °C, sample weight approximately 15 mg.

2.2.7. Inductively coupled plasma optical emission spectroscopy (ICP-OES)

The concentrations of elements of Li, Ni, Mn and Co were determined by the inductively coupled plasma emission spectrometer 5900 ICP-OES (Agilent, USA). The plasma was generated from the Ar flow using a water-cooled semiconductor radiofrequency generator with a frequency of 27 Hz and a power of 1.2 kW. The measurements were performed in the so-called synchronous vertical dual view (SVDV) mode, which allows measurements in axial and radial views simultaneously. Quantitative analysis was based on a five-point calibration series for each measured element, ranging from 0.2 to 1.0 mg/L. The results are based on three measurements of each sample.

3. Results

3.1. Composition and structure

Composition and structure were monitored using XRF and XRD on all samples from the thermally untreated sample (NMC0) to the sample annealed to the highest temperature investigated (NMC401). For comparison, SEM/EDS analysis was also conducted on the same set of samples.

The XRF analysis of the chemical composition (see Table 1) of the untreated sample NMC0 identified nickel oxide (56.2 wt%), manganese oxide (13.4 wt%) and cobalt oxide (11.7 wt%) as the major elements. Other minor elements were aluminium, which corresponds to the material from which the current collector is made, phosphorus, and fluorine, which come from other parts of the battery, such as electrolytes or binder used for the cathode material. Another identified minor element was tungsten. It is likely a dopant in the form of, for example, tungsten

Table 1
Chemical composition in wt.% analysed by XRF in the reference material (NMC0) and in the heat-treated materials (NMC101-401).

Analysed composition [wt. %]	Sample				
	NMC0	NMC101	NMC201	NMC301	NMC401
Al_2O_3	0.6 ± 0.0	1.6 ± 0.0	1.0 ± 0.0	0.8 ± 0.0	7.0 ± 0.1
P_2O_5	2.3 ± 0.1	1.5 ± 0.0	2.3 ± 0.0	1.5 ± 0.0	1.0 ± 0.0
F	15.2 ± 0.6	9.5 ± 0.5	15.7 ± 0.6	8.9 ± 0.5	2.9 ± 0.2
MnO	13.4 ± 0.1	14.3 ± 0.1	13.4 ± 0.1	14.4 ± 0.1	14.4 ± 0.1
Co_3O_4	11.7 ± 0.1	12.5 ± 0.1	11.5 ± 0.1	12.6 ± 0.1	12.9 ± 0.1
NiO	56.2 ± 0.2	60.0 ± 0.2	55.5 ± 0.2	61.3 ± 0.2	61.3 ± 0.1
WO_3	0.6 ± 0.0	0.6 ± 0.0	0.7 ± 0.0	0.5 ± 0.0	0.4 ± 0.0

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oxide (WO_3), aimed at improving electron conductivity and enhancing the kinetics of redox reactions in cathode materials [30].

Some changes in composition, particularly in the content of minor compounds, occur as a result of heat exposure. Specifically, a decrease in the content of P_2O_5, F, and WO_3 is observed, while the content of Al_2O_3, MnO, Co_3O_4, and NiO increases.

The increase in Al_2O_3 and the decrease in P_2O_5 and F can be attributed to the thermal treatment process of the PVDF-based polymer binder in the battery electrolyte. During heat treatment, fluorine and phosphorus components are prone to evaporate or undergo chemical transformations, leading to a reduction in their presence within the material. Since the composition analysis is always recalculated to 100 %, any decrease in F and P_2O_5 will naturally result in an increase in other components, including Al_2O_3. However, it is also possible that the increased content of Al_2O_3 could be attributed to the precipitation of Al compounds on the NMC particles, as discussed in Ref. [32]. It is conceivable that during the separation process of NMC particles from Al foil in demineralized water, this phenomenon occurred. The increased content of F in the NMC201 sample is consistent with the results of the XRD analysis (Fig. 2d), where the crystalline phase Li_2NiF_4 is observed.

The SEM/EDS results (see Table 2) are consistent with the findings obtained from XRF. They confirm a decrease in the content of minor components of the material, accompanied by an increase in Ni, Mn, and Co element. This indicates that the organic components of the material and compounds from the PVDF binder and electrolyte evaporate with increasing temperature. The EDS analysis results showed a significant decrease in F content, particularly between samples exposed to 300 °C (NMC301) and 400 °C (NMC401). The majority of impurities from other battery components evaporate between these temperatures, as evidenced by the sharp decline in C content.

Fig. 1 presents images obtained using a scanning electron microscope of the cathode material surface. On the left side of each subfigure, the images were captured in secondary electron (SE) mode. On the right side, EDS maps of the elements Ni, Mn, Co, and F are displayed. No significant changes were observed between the samples, except that the EDS maps indicate a decrease in fluorine content with increasing annealing temperature. This suggests a reduction in the polymer binder as the temperature rises.

X-ray diffraction analysis (see Fig. 2) confirmed the presence of the crystalline phase $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ (ICDD 01-084-9844) in all samples, corresponding to the NMC622 composition. The results indicate that heat treatment of the samples in the selected temperature range of 100–400 °C does not affect the material's structure, except for the sample heated to 200 °C (NMC201). At this temperature, the phase Li_2NiF_4 was identified, suggesting a structural change in the material. This phase likely originates from either the reaction between cathode material and the electrolyte or from the decomposition of PVDF. All identified peaks were properly labelled in the diffractogram. Subsequent chemical analysis, as detailed in Table 1, revealed NiO, MnO, and Co_3O_4 as predominant oxides, with NMC0 exhibiting a relatively elevated fluorine content compared to the NMC401 specimen. This disparity aligns with the characteristics of the binder used, which exhibits a higher melting point. Other elements or oxides were present only in marginal proportions.

Table 2
The chemical composition of the cathode material at different thermal exposures performed by EDS, given in wt.%.

Element	Sample				
	NMC0	NMC101	NMC201	NMC301	NMC401
Al	0.5 ± 0.0	0.9 ± 0.1	0.7 ± 0.0	0.4 ± 0.0	0.9 ± 0.1
F	19.9 ± 0.1	20.6 ± 0.5	20.7 ± 0.2	20.8 ± 0.1	3.0 ± 0.3
Ni	30.2 ± 0.1	26.9 ± 1.1	29.6 ± 0.2	32.0 ± 0.2	47.9 ± 1.7
Mn	9.4 ± 0.1	9.1 ± 0.4	9.2 ± 0.1	9.9 ± 0.1	14.2 ± 0.8
Co	7.2 ± 0.1	6.8 ± 0.6	7.0 ± 0.1	7.7 ± 0.1	12.3 ± 1.1
W	1.2 ± 0.2	1.4 ± 0.2	1.0 ± 0.2	1.0 ± 0.2	0.4 ± 0.2
P	0.6 ± 0.0	1.4 ± 0.1	0.60 ± 0.0	0.7 ± 0.0	0.8 ± 0.1
C	9.0 ± 0.2	9.0 ± 1.4	9.3 ± 0.2	8.7 ± 0.2	3.7 ± 0.4
O	22.1 ± 0.1	23.8 ± 0.6	22.0 ± 0.1	18.8 ± 0.1	16.9 ± 0.6

3.2. Thermal analysis

Thermogravimetric measurement of sample NMC0 in air atmosphere (Fig. 3) showed a small weight loss up to 230 °C which is attributed to electrolyte loss. The decomposition between 230 and 630 °C took place in two main steps with peaks at 270 and 540 °C; the loss in this interval is mainly attributed to the decomposition of the present polymers. Significant thermooxidation, followed by mass loss, was observed at temperatures above 750 °C. Most of the observed changes occur outside our studied temperature range (above 400 °C).

3.3. Concentrations of Li, Ni, Mn, and Co

The results of the ICP-OES analysis showed that the measured concentration depends on the dissolution time, the type of solvent, and the temperature to which the initial material was exposed. In a 20 % HNO_3 solution (see Fig. 4), the measured concentrations were several times higher than in demineralized water. In terms of the dependence of the measured concentration on the dissolution time, the highest values were measured after 4 h. For the NMC101 and NMC301 samples, a significant decrease in concentration was observed after 2 h of dissolution, indicating that precipitation of the determined ions occurred in the solution.

The temperature to which the initial material was exposed before dissolution also plays an important role, as can be seen from the graphs, the concentrations of Ni, Mn, and Co are significantly higher in the case of material exposed to temperatures of 300 and 400 °C, respectively. This phenomenon is likely associated with the melting temperature of polyvinylidene fluoride (PVDF), the binder used in the manufacture of these batteries, which could affect the thermal stability and performance of the battery cells. Nickel was the most leached element from the NMC material, followed by manganese, cobalt, and the lowest values were measured for lithium. The mean relative standard deviations were 0.24 % for lithium, 0.31 % for nickel, 0.42 % for manganese and 0.39 % for cobalt.

Zhang et al. described how PVDF affects the dissolution of the cathode material after thermal treatment due to its semicrystalline nature that allows the electrolyte to contact the active material directly, increasing dissolution and solubility in solutions. Additionally, the PVDF polymer primarily relies on weaker van der Waals forces, leading to significant thermal expansion. This large volume expansion can increase the internal pressure of the battery, further exposing the active material to the electrolyte, contributing to higher solubility, performance deterioration, and safety issues [35].

Demineralized water was chosen as the second type of solvent because it is much safer to use than acids. After dissolution in demineralized water, only lithium concentrations were measured (see Fig. 5), which were almost constant after 2 h of dissolution. For the other elements, it would be appropriate to choose a different procedure or solvent type for quantitative analysis due to the values that were below the detection limit of the instrument. The mean relative standard deviation was 0.31 %.

3.4. Theoretical valuable metal yield

To determine the yield, the maximal theoretical amount was computed utilising the technical datasheet specifications. The cathode material composes 42.5 % of a cell. Therefore from

$$m_{cathode} = m_{pack} \bullet \frac{p}{100} \quad (2)$$

where p is a portion of the active cathode material, and the cathode mass of a cell is

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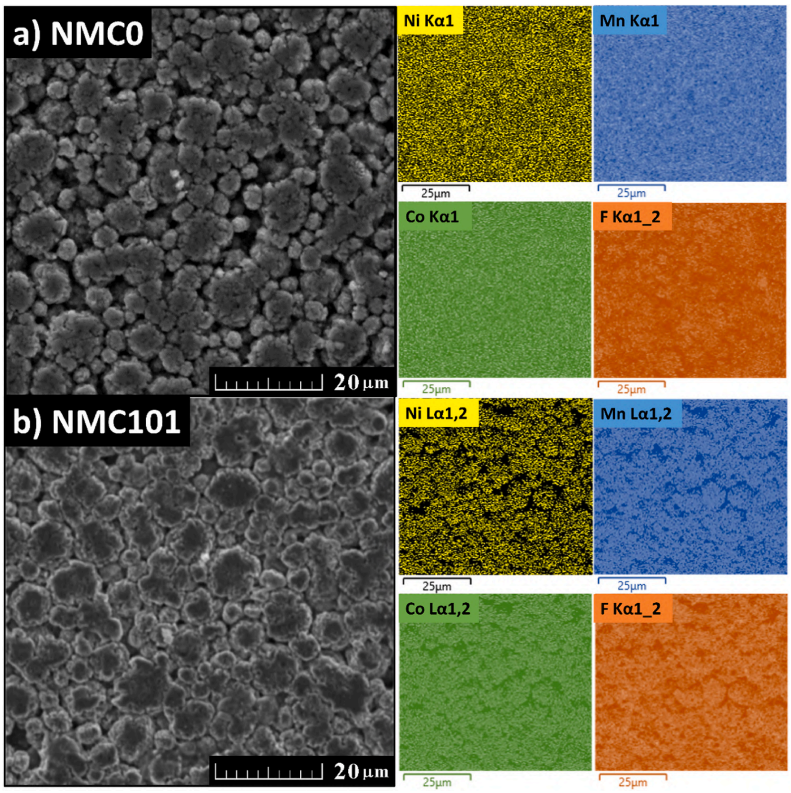

Fig. 1. Scanning electron microscopy and energy dispersive spectroscopy of the cathode material surface for samples (a) NMC0, (b) NMC101, (c) NMC201, (d) NMC301 and (e) NMC401.

$$M_{cathode} = 0.458\ kg \tag{3}$$

The mole and mass quantities of individual elements of $Li_{1.0}Ni_{0.6}Mn_{0.2}Co_{0.2}O_2$ were calculated based on formula

$$n_A = u_A \bullet \frac{m_{cathode}}{M_{cathode}} \tag{4}$$

where n_A is the number of moles of the respective element, u_A is the stoichiometric coefficient [4]. From the mass concentration measured by ICP-OES, the mass of dissolved ions was calculated for the highest measured concentrations, in this case for samples dissolved for 4 h (see Table 3a). From this mass, the theoretical yield of valuable metals obtained by dissolution in 20 % nitric acid solution and demineralized water, respectively, was calculated. The results shown (see Table 3b) are for the NMC401 sample after 4 h of dissolution.

The masses of dissolved valuable metals increase with increasing annealing temperature of the sample. The highest results are obtained for the sample annealed at 400 °C. The dissolution in nitric acid gave a lithium yield almost five times higher than that obtained in demineralized water.

The efficiency of this method was compared with the available results of the hydrometallurgical method. The highest recovery (91 %) was found for nickel; the recovery of the other elements studied ranged from 60 to 70 % using acid. In demineralized water, the highest lithium recovery was 14 %.

4. Discussion

The cathode sample was subjected to thermal exposure and subsequently dissolved in appropriate solvents to simulate the initial stages of the pyrometallurgical-hydrometallurgical process. Annealing temperatures of 100, 200, 300 and 400 °C were applied for 1 h each, and dissolution was carried out for 1.5, 2 and 4 h. Changes after both thermal treatment and dissolution in the specified solvents were closely monitored.

Chemical composition analysis revealed a reduction in the fluorine and phosphorus content with an increase in the temperature to which the material was exposed. This suggests that, as the temperature rises, the binder undergoes decomposition and any residual electrolyte adhering to the cathode samples is burnt off. The XRD results indicate that the material composition remains NMC622 after exposure to heat.

Examination of the samples using an SEM/EDS instrument confirmed the loss of binder with increasing temperature, as depicted in the attached photographs. Furthermore, consistent trends in chemical composition were observed, as observed in the X-ray fluorescence

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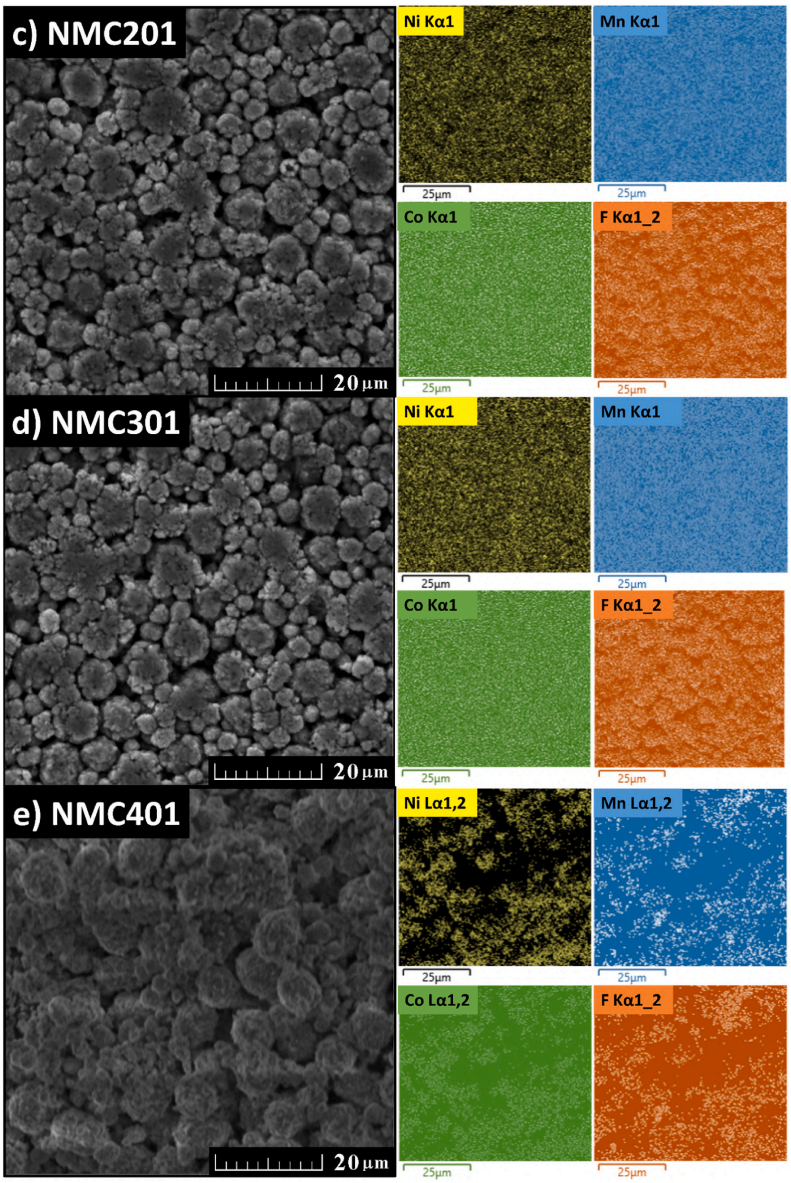

Fig. 1. (*continued*).

results - specifically, a decrease in the content of fluorine and phosphorus in the case of sample NMC401.

The TGA results confirmed that the binder decomposition occurs within the temperature range of 230–630 °C. This discovery implies that the material can undergo heat treatment at lower temperatures, such as 400 °C, allowing reutilization of the active material. At this temperature, most of the binder has already undergone decomposition, while the structure of the NMC remains intact. This not only minimises recycling expenses, but also ensures the feasibility of material reuse.

Measurements from ICP-OES indicated that the concentration of lithium, nickel, manganese, and cobalt is dependent on the dissolution time and, notably, on the temperature to which the initial material was

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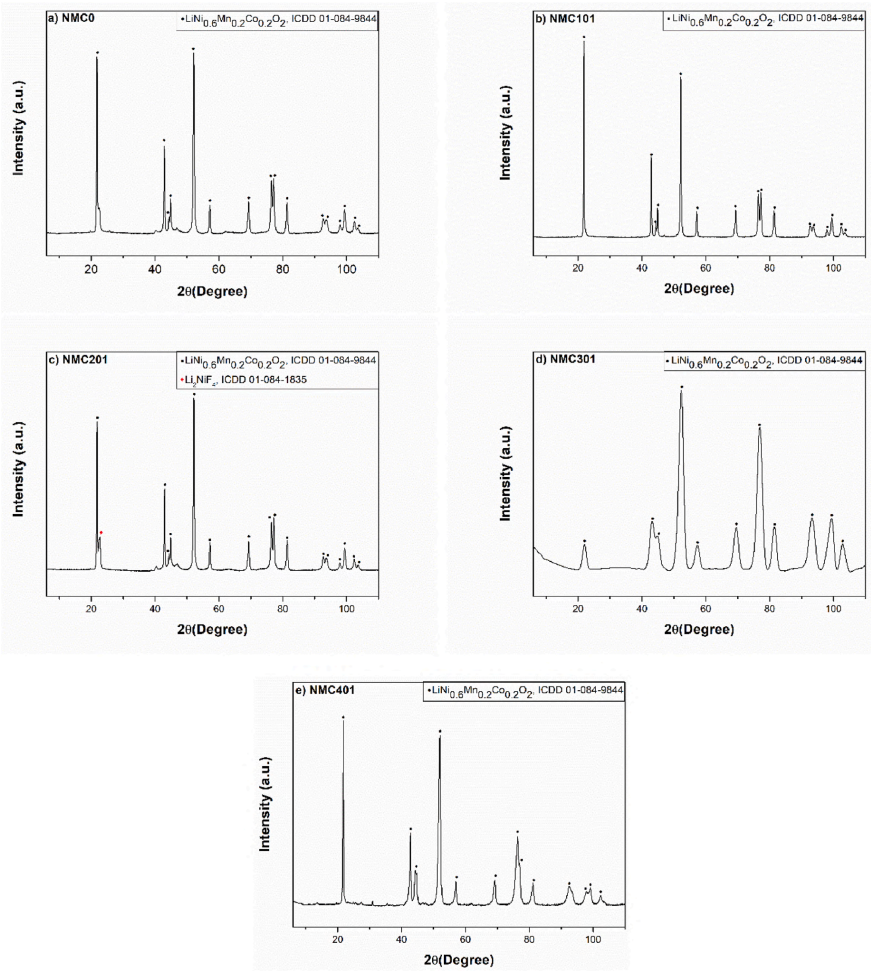

Fig. 2. XRD analysis of NMC0-NMC401.

exposed. As the temperature increased, the concentration of all elements also increased, with the highest concentration observed in the case of sample NMC401. This further supports the notion that annealing decomposed the binder, leading to the release of a higher quantity of ions into the solution from the sample subjected to the highest temperature. From the measured concentrations, the masses of dissolved ions were calculated and from these the efficiency of the process was calculated and compared to the efficiency of metal recovery from spent NMC using the hydrometallurgical method, which is the most similar in principle to our method due to the low temperatures used in our study. The theoretical efficiency of our method was similar in the case of nickel recovery (we achieved 91 %), while it was lower for the other elements investigated. This is probably due to the use of a different solvent. In the compared study, sulfuric acid was used in combination with hydrogen peroxide [36]; in this work, nitric acid was used as the solvent. Although the lithium yield obtained by dissolving the material in demineralized water was fourteen times higher than in the other reported study [32], dissolution in acid gave much higher results. The higher yield obtained in our study is probably due to the longer dissolution time; on the other hand, most of the lithium is released into solution at the beginning of the process, so the heat treatment of the sample, which was carried out in this paper, will have a great influence here. Due to the highest yields at the highest temperature investigated, this work will be followed by the investigation of material fired at higher temperatures, and therefore there is the potential for further release of the binder at higher temperatures, and therefore higher yields.

5. Conclusions

Based on the results of the analyses, the cathode material retains its structure even when exposed to temperatures of 100–400 °C. XRD confirmed the structure of NMC622 even after annealing the material at

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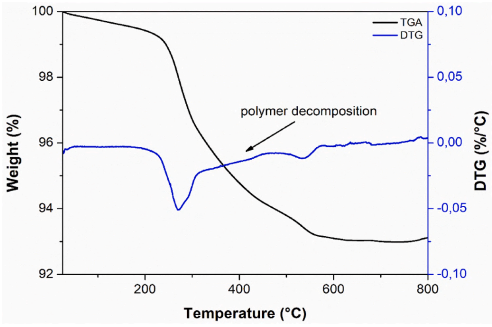

Fig. 3. TGA curve of the sample NMC0.

the studied temperatures (100–400 °C). However, decomposition of the binder or leaching of electrolyte residues occurs in this temperature range, and therefore changes in composition were observed (XRF, EDS). The effect of annealing temperature is most clearly observed in the graphs showing the dependence of concentration on dissolution time, with sample NMC401 reaching the highest measured concentrations, and always after 4 h of dissolution. From these concentrations, the theoretical valuable metal yield was calculated, which is highest at the highest temperature investigated. From this it can be said that annealing at 400 °C for 1 h followed by dissolution in 20 % nitric acid for 4 h leads to the highest yield. The efficiency of the process was 91 % for nickel, while for the other metals studied it was in the range of 60–70 %, which is a lower yield compared to other methods; however, nitric acid, as the solvent used is cheaper compared to other solvents used. At the same time, e.g., the pyrometallurgical method involves smelting of the material at very high temperatures (around 1500 °C), so our method could significantly reduce both the economic and environmental associated with the pre-treatment of the material. A comprehensive evaluation that compares our method with conventional recycling pre-treatment techniques and an analysis of the potential economic benefits is intended for future research. By dissolving in demineralized water, an efficiency of around 14 % was achieved, which is much higher than in another study where around 1 % was achieved. Therefore, dissolution in acid is more efficient in terms of yield; on the other hand, dissolution in water is a highly safe and more environmentally friendly. Since the highest yields of valuable metals were found at the highest observed annealing temperature, as well as binder decomposition, and it is therefore possible that complete binder decomposition occurs at higher temperatures, a follow-up experiment will be developed to investigate the behaviour of the same material at higher temperatures, specifically in the 500–800 °C range.

CRediT authorship contribution statement

Nikola Klusoňová: Writing – original draft, Investigation, Writing – original draft, Investigation. **Eliška Sedláčková:** Writing – review & editing, Investigation, Writing – review & editing, Investigation. **Jan Kočí:** Data curation, Conceptualization, Data curation, Conceptualization. **Dominik Pilnaj:** Methodology, Data curation. **Karolína Pánová:** Writing – review & editing, Formal analysis, Data curation. **Jonáš Uřičář:** Writing – review & editing, Formal analysis, Data curation, Writing – review & editing, Formal analysis, Data curation. **Václav Procházka:** Writing – review & editing, Formal analysis, Writing – review & editing, Formal analysis. **Kristýna Jílková:** Writing – review & editing, Project administration, Conceptualization. **Anna Pražanová:** Writing – review & editing, Resources, Conceptualization. **Martin**

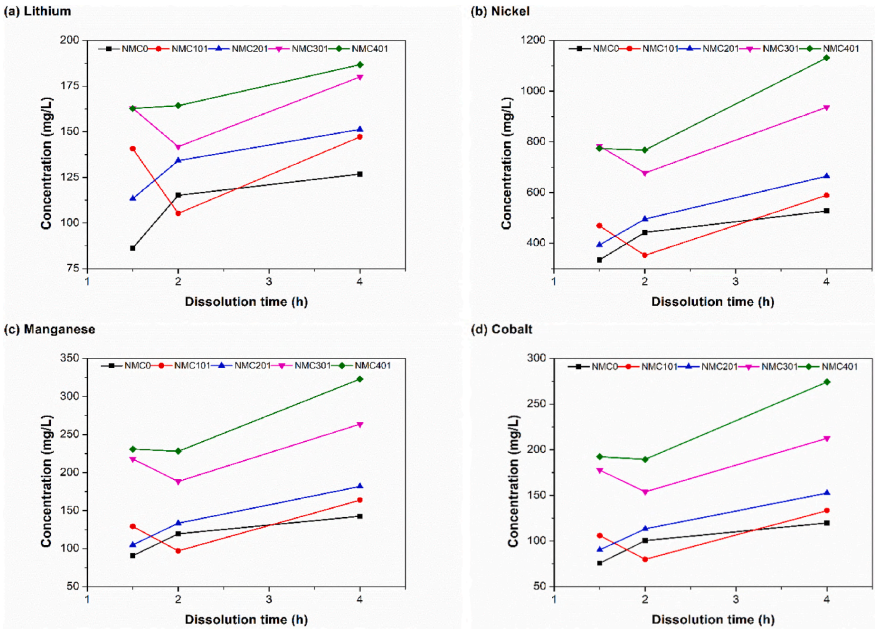

Fig. 4. Concentration of (a) lithium; (b) nickel; (c) manganese and (d) cobalt dissolved in 20 % HNO_3.

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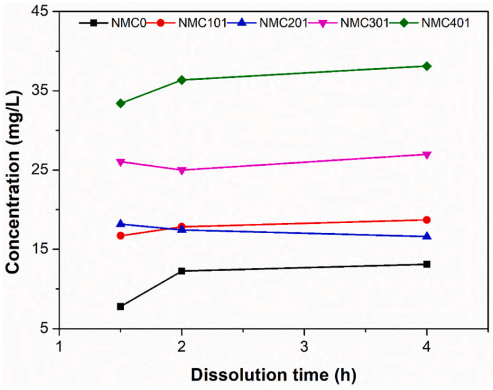

Fig. 5. Concentration of lithium dissolved in demineralized water.

Table 3
(a) Mass [16] of valuable metals from a sample (100 mg) determined from the measured concentration after 4 h of dissolution; (b) Comparison of the efficiency [%] of the hydrometallurgical method and with the results of our work.

(a)

	Our study				
	NMC0	NMC101	NMC201	NMC301	NMC401
Lithium in HNO_3	3.2	3.7	3.8	4.5	4.7
Lithium in demin. water	0.3	0.5	0.4	0.7	1.0
Nickel in HNO_3	13.2	14.7	16.6	23.4	28.3
Manganese in HNO_3	3.6	4.1	4.5	6.6	8.1
Cobalt in HNO_3	3.0	3.3	3.8	5.3	6.9

(b)

Methods	Leaching agent	Leaching condition	Efficiency [%]	Ref.
Hydrometallurgy	$H_2SO_4 + H_2O_2$	40 °C, 1 h	Li, Ni, Mn, Co = 99.7	[36]
	deionised water	room temperature 30–60 min	Li = 1.0	[32]
Our study	HNO_3	pre-treated at 400 °C room temperature 4 h	Li = 69.9, Ni = 91.0, Mn = 63.6, Co = 60.4	–
	demineralized water	pre-treated at 400 °C room temperature 4 h	Li = 14.4	–

Havlík Míka: Supervision.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Nikola Klusonova reports financial support was provided by University of Chemistry and Technology Prague. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

I have shared the link to my data at the Attach File step.
Dataset of "Thermal Stability of Valuable Metals in Lithium-Ion Battery Cathode Materials: Temperature Range 100-400 °C" (Original data) (Zenodo)

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batteries

MDPI

Review

Direct Recycling Technology for Spent Lithium-Ion Batteries: Limitations of Current Implementation

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Abstract: The significant deployment of lithium-ion batteries (LIBs) within a wide application field covering small consumer electronics, light and heavy means of transport, such as e-bikes, e-scooters, and electric vehicles (EVs), or energy storage stationary systems will inevitably lead to generating notable amounts of spent batteries in the coming years. Considering the environmental perspective, material resource sustainability, and terms of the circular economy, recycling represents a highly prospective strategy for LIB end-of-life (EOL) management. In contrast with traditional, large-scale, implemented recycling methods, such as pyrometallurgy or hydrometallurgy, direct recycling technology constitutes a promising solution for LIB EOL treatment with outstanding environmental benefits, including reduction of energy consumption and emission footprint, and weighty economic viability. This work comprehensively assesses the limitations and challenges of state-of-the-art, implemented direct recycling methods for spent LIB cathode and anode material treatment. The introduced approaches include solid-state sintering, electrochemical relithiation in organic and aqueous electrolytes, and ionothermal, solution, and eutectic relithiation methods. Since most direct recycling techniques are still being developed and implemented primarily on a laboratory scale, this review identifies and discusses potential areas for optimization to facilitate forthcoming large-scale industrial implementation.

Keywords: lithium-ion batteries; direct recycling; end-of-life

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1. Introduction

Lithium-ion batteries (LIBs) stand out as a pivotal technology with numerous advantages compared to other electrochemical storage technologies these days. These include higher cell voltage, increased energy density, extended lifespan, minimal memory effect, simplicity to charge and maintain, low self-discharge, potential for miniaturization, and different packaging shapes. Thus, the LIBs set a turning point in the field of power sources, where they are utilized in a variety of applications encompassing small consumer electronic systems, including watches, smartphones, or laptops; portable power tools, such as drills or saws; home or industrial systems for energy storage (BESS); and light or heavy means of transport, including electric bikes, scooters, and electric vehicles (EVs) [1–4].

The typical composition of LIBs predominantly comprises a cathode, anode, separator, and electrolyte. Ordinarily, batteries also have an electronic control unit, a protective metal casing, and plastic coverings according to shape, composition, and structure, as illustrated in Figure 1 [5]. Cathodes are predominantly composed of a mixture involving a polyvinylidene fluoride (PVDF) binder, carbon powder, and lithium (Li) transition metal oxides such as lithium cobalt oxide (LCO or $LiCoO_2$), lithium manganese oxide (LMO or $LiMn_2O_4$), lithium iron phosphate (LFP or $LiFePO_4$), lithium nickel cobalt aluminum oxide (NCA or $LiNi_xCo_yAl_zO_2$), and lithium nickel manganese cobalt oxide (NMC or $LiNi_xCo_yMn_zO_2$). These materials are coated onto a current collector, typically an aluminum (Al) foil. Similarly, anodes commonly utilize graphite, providing a structure capable of holding Li within

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its layers. The active carbon material is attached to a copper (Cu) conductor plate using a PVDF binder. Electrolytes consist of a solution containing a lithium-containing salt, such as lithium perchlorate ($LiClO_4$), lithium tetrafluoroborate ($LiBF_4$), or lithium hexafluorophosphate ($LiPF_6$), dissolved in an organic solvent like dimethyl carbonate (DMC) or ethylene carbonate (EC). As a medium, the electrolyte facilitates the transport of ions between battery electrodes. A separator, crafted from polymeric materials such as polypropylene (PP) or polyethylene (PE), acts as a non-conductor or barrier; thus, it is inserted between the anode and cathode to eliminate the risk of short-circuiting caused by direct contact between the electrodes [1,6,7]. The composition of LIBs slightly differs between manufacturers and commonly consists of 5–20% Co, 5–20% Mn, 5–30% Ni, 5–10% Li, 5–40% of other metals, 10–15% organic chemicals, and 3–10% plastics [8–10].

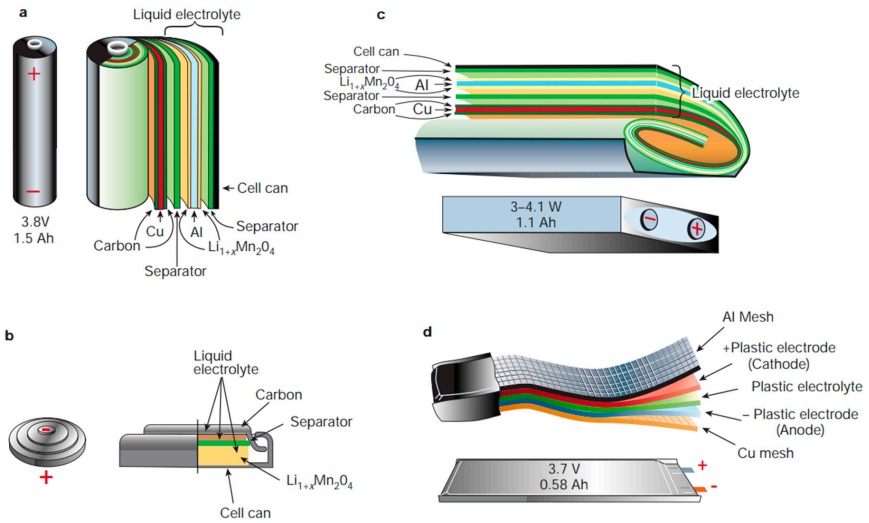

Figure 1. Schematic drawing showing the shape and components of various Li-ion battery configurations: (**a**) cylindrical; (**b**) coin; (**c**) prismatic; and (**d**) thin and flat. Reprinted from [5], with permission from Springer Nature, Copyright (2001).

The lifespan of LIBs varies according to their application field; in a small electronic device, it can last between 2 and 5 years, whereas in EVs or BESSs, it may range from 8 to 15 years [1,11]. Considering the significant difference between the utilization of LIB within small portable electronic devices and longer-life applications, currently standing at an 80:20 ratio, the annual increase in end-of-life (EOL) batteries is evident [12]. Projections of trends have become a focal point in numerous predictions; according to them, the estimation of spent LIBs will increase to 464,000 tonnes by 2025 [13]. In response to the challenges of the battery raw material supply chain and environmental footprint reduction, a series of global or European initiatives have been implemented. The simplified hierarchy for EOL battery management is outlined as follows [1,14]:

- **Prevention:** designing LIBs with less-critical materials and creating electronic devices with smaller/lighter LIBs to prevent and minimize waste generation.
- **Re-use:** entailing secondary usage of LIBs to prolong their service life, especially for EV batteries.
- **Recycling:** involving the recovery of valuable materials from spent LIBs and their return to the value chain.

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- **Recovery:** considering using certain materials from spent LIBs as fuel in processes like pyrometallurgy to extract energy from waste.
- **Disposal:** discarding spent LIBs without recovered value, directing them to specialized landfills or municipal waste combustion facilities for incineration.

Given the LIBs' toxic content, which can cause severe environmental pollution risks and pose a threat to human health, and the significant quantities of valuable and other essential metals, such as Li, Co, Ni, Mn, Cu, Al, and Fe, recycling emerges as an up-and-coming technology for managing waste LIBs. This approach, covering suitable flue gas treatment, mitigates the pollution from hazardous materials and offers alternative sources of essential metals, particularly Li and Co, while reducing emissions associated with mining and post-processing activities [2,15].

Commonly implemented methods to recycle appealing components of spent LIBs are either based on pyrometallurgical or hydrometallurgical methods. The pyrometallurgical procedure, rooted in metal smelting, is a well-established practice based on separation and high-temperature treatment [16]. While the process is energy-intensive and emission-productive, it proves relatively efficient as it yields the most valuable metal resources (e.g., Co or Ni) via metal alloys. However, portions of Li end up in slags, which are not typically recovered [17]. Conversely, hydrometallurgical techniques allow the recovery of all metal elements in their salt states with a higher purity [17,18]. This approach serves at the expense of consuming significant chemical reagents, resulting in elevated costs and environmental concerns related to waste pollution [1,2,19].

These methods represent quick and sufficient solutions for current recycling pressure, but they cannot realize a closed-loop economy in future years without the necessary optimization steps. Therefore, there is a high demand for alternative recycling technologies that can directly reuse degraded energy storage materials for battery manufacturing from an economically and environmentally sustainable perspective. The most promising of them is direct recycling technology [7,20,21].

Lately, direct recycling processes have surfaced as a sustainable recycling technology renowned for healing compositional and structural defects [7]. By preserving the original crystal structure and avoiding the breakdown of electroactive materials into their elemental states, it contrasts conventional recycling methods (pyrometallurgy, hydrometallurgy) [2]. A comparison of the general pros, cons, and challenges of the mentioned recycling processes is shown in Table 1.

Although hydrometallurgy is known for its high process efficiency, which can reach up to 76–98.2%, and the purity of obtained materials between 96.5–99.7%, low energy consumption is typically only observed during the recovery phase [2]. However, when considering additional processes such as wastewater treatment, energy consumption can equal or surpass that of pyrometallurgy [22]. Due to the inherent nature of hydrometallurgical processing, this method requires more water than other techniques. Thus, optimized approaches that minimize water usage can yield significant additional benefits for this recycling method [20].

Despite the advantages of pyrometallurgy, which include process simplicity and speed compared to multi-step chemical leaching processes that span several hours, the energy consumption during the recovery phase is notably high due to the requirement of operating at high temperatures [20]. Moreover, the process leads to significant emission production. Thanks to high-thermal processing, recovering certain materials like Li, Al, and organic compounds is impossible. Although pyrometallurgical processing does not restrict input size, it cannot be used to treat some LIB types, such as LFP. Therefore, the method is often combined with hydrometallurgical recycling steps to enhance efficiency and product quality [2].

Consequently, direct recovery holds considerable promise in delivering recycled electroactive materials to battery manufacturers with heightened sustainability, characterized by decreased energy consumption and limited dependence on chemical agents [20,21].

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Nevertheless, it is currently necessary to optimize established techniques to reduce costs and further optimize product performance.

Table 1. Comparisons of advantages and disadvantages and challenges for different methods. Reprinted from [23] under the terms and conditions of the CC BY 4.0 license, Copyright (2020).

Process	Advantages	Disadvantages	Challenge
Hydrometallurgical process	High recovery rate High-purity product Low energy consumption Less waste gas High selectivity	More wastewater Long process	Wastewater treatment Optimize the process
Pyrometallurgical process	Simple operation and short flow No requirement for categories or size of inputs High efficiency	Li and Mn are not recovered High energy consumption Low recovery efficiency More waste gas and the cost of waste gas treatment	Reduce energy consumption and pollution emissions Reduce environ. hazards Combine hydrometallurgy well
Direct recycling process	Short recovery route Low energy consumption Environmentally friendly High recovery rate	High operation and equipment requirements Incomplete recovery	Reduce recovery costs Lower the requirements for categories Further optimize product performance

This work examines the current direct recycling procedures for spent LIB materials, including common cathode (NMC, LFP, LCO, and LMO) and graphite anode treatment. The review introduces the technical challenges, limitations, and barriers of five different direct recycling methods (solid-state sintering, electrochemical, ionothermal, solution, and eutectic relithiation), as illustrated in Figure 2. Although these methods provide high efficiency in restoring the properties of regenerated materials to a level comparable to pristine, the critical challenge in direct recycling revolves around the processing of black mass and the necessity of recycling pre-treatment within the EOL process scenario. Thus, this work discusses potential optimization options that could lead to the industrially scaled implementation of direct recycling.

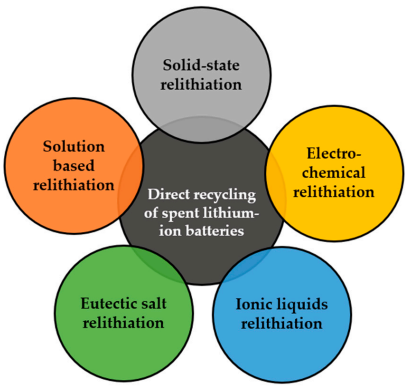

Figure 2. Overview of the current direct recycling approach, covering the relithiation techniques.

2. Direct Recycling Technology

In the realm of recycling spent LIBs, the technological processes are divided into two separate routes: recycling pre-treatment and the subsequent material recovery, i.e., the recycling procedure [2]. The battery pre-treatment precedes further recycling treatment and can improve the efficiency of valuable material recovery, process safety, or lower

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energy consumption. These processes primarily separate the active electrode materials from the current buses, separators, electrolytes, and covers or connections. Presently, the pre-treatment process lacks a well-defined design, leading to the adoption of various approaches in recent years. Typically, the process can be outlined through the following seven-step procedure; the short-cuts of the individual steps' details are given in Table 2, and the review work by Kim et al. [19] provides a comprehensive description.

- Discharging;
- Dismantling;
- Comminution;
- Classification;
- Separation;
- Dissolution;
- Thermal treatment.

The main aim of the recycling pre-treatment phase is to eliminate impurities and achieve the highest possible quality in the output product, the so-called "black mass". This finely ground fraction predominantly comprises cathode materials (cathode black mass, also known as CBM) and serves as the input material for the subsequent stages of recycling processing [2,22].

Table 2. Summary of recycling pre-treatment process steps [2,19].

Recycling Pre-Treatment Step	Step Characterization
Discharging	Discharge systems with electric (active or passive) loads, deep discharging < 1 V.
Dismantling	Manual disassembly from battery (pack) to the module or cell level.
Comminution	Crushing active electrode materials into fine fractions, implemented by dry or wet procedures.
Classification	Categorization according to the fraction product size, typically applies by sieving.
Separation	Specialized separation methods, including magnetic, eddy current, or electrostatic techniques.
Dissolution	Chemical separation of active cathode materials from Al foils.
Thermal treatment	Complete removal of the organic parts, such as binders, additives, plastics, or carbon black. Final product: black mass.

Following pre-treatment, the black mass, primarily comprising the gathered spent cathode materials, undergoes regeneration through either pyrometallurgical, hydrometallurgical, or direct recycling methods. A complex overview of the implemented pyrometallurgy or hydrometallurgy techniques, including a detailed examination of their advantages and disadvantages, along with considerations of economic and environmental perspectives, can be found in works by Pražanová et al. [20], or Lander et al. [24].

Regarding direct recovery pathways, the CBM is set for regeneration, encompassing the restoration of the crystal structure while concurrently mitigating the loss of Li electroactivity at its original location—a process commonly referred to as "in situ" electroactivity offsetting. This healing methodology is introduced in response to the common issues of inactive Li supply and the degradation of battery performance resulting from irreversible structural transitions, such as the formation of the solid electrolyte interphase (SEI) layer on the anode surface; changes in volume and crystal phase; structural stress; mechanical strains; and micro-cracks between/in the particles. A detailed description of the struc-

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tural changes within the LIB cells, which lead to a performance decrease and Li losses, is presented in work by Wu's [22] and Xu's [7] research groups.

Thus, direct recycling emerges as a promising technique within currently implemented recycling methodologies. It possesses considerable potential for reinstating the electrochemical performance of cathode materials and their rejuvenation to reach quality levels similar to those of fresh materials. This regeneration involves restoring entire structures, not the recovery of individual valuable metals, as illustrated in Figure 3. A notable advantage of this method is that it reduces environmental impacts and lowers economic costs [22].

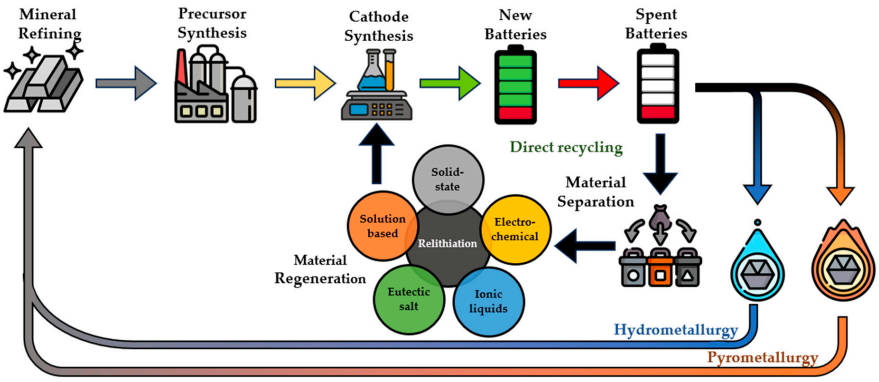

Figure 3. Example of the main direct recycling steps; comparison with pyro-/hydrometallurgy [22].

The example of the cost, energy consumption, and environmental impacts comparison of producing 1 kg of NMC111 cathode (nickel manganese cobalt oxide in a 1:1:1 mass ratio for Ni:Mn:Co) and recycling them with different technologies (namely, pyro-/hydrometallurgy and direct recovery) is shown in Figure 4 [22].

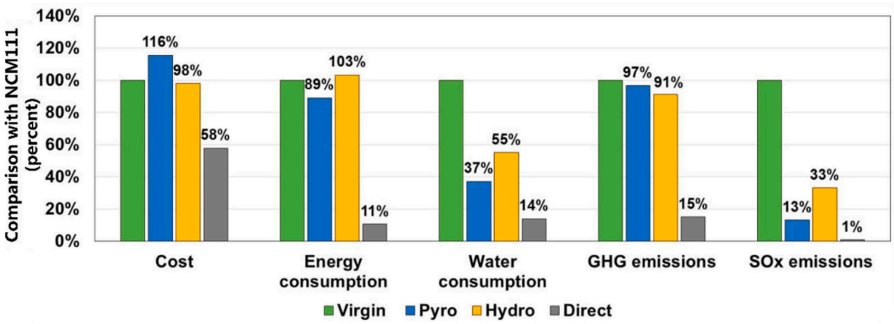

Figure 4. The comparison of the cost, energy consumption, and environmental impacts of producing 1 kg of NMC111 cathode and recycling it with different technologies (namely, pyro-/hydrometallurgy and direct recovery). Reprinted from [22], with permission from Elsevier, Copyright (2023).

However, for quality and highly representative results, evaluating each established process individually with all its technological aspects or additional steps is necessary. While the work by Wu et al. [22] lacks specific compared-process details, the research-based trends can be observed through selected categories [20,25–28]. Generally, hydrometallurgy exhibits the highest energy consumption due to its intricate and energetically demanding steps, particularly in the purification of waste products, notably water. Consequently,

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water consumption is the most significant via hydrometallurgical processes. Conversely, pyrometallurgy, characterized by high-temperature processing, contributes significantly to greenhouse gas (GHG) emissions, although sulfur oxide (SO_x) emissions predominantly stem from chemical (hydrometallurgical) processing. Economic evaluations commonly depend on the accuracy of process characterizations. However, given the high technology costs associated with high-temperature processing, pyrometallurgical processes are presumed to incur substantial expenses in terms of investment and operation. Moreover, this comparison underscores the primary objectives of direct recycling, which entail low operating costs and minimal environmental impact.

The direct recycling process can be generally called "relithiation" [29]. These approaches mainly include solid-state sintering, electrochemical relithiation in organic and aqueous electrolytes, ionothermal relithiation, solution relithiation, and eutectic relithiation, whose advantages and challenges are summarized in Table 3 [7].

Table 3. The advantages and challenges of different direct recycling technologies. Reprinted from [7], with permission from Wiley-VCH GmbH, Copyright (2023).

Direct Recycling Method	Advantages	Challenges
Solid-state relithiation	One-step regeneration; Simple operation	Pre-determination of Li deficiencies; Phase impurity
Electrochemical relithiation	Low energy consumption; Low cost	Pre-determination of Li deficiencies; Removal of current collector, binders, . . . Phase impurity; Scalability issue
Ionic liquids relithiation	Self-saturation relithiation	High cost of ionic liquid; Scalability issue
Eutectic salt relithiation	Self-saturation relithiation; Low energy consumption	Removal of solidified salts; mixture after regeneration; Scalability issue
Solution based relithiation	Self-saturation relithiation; Wide applicability; Low cost	Safety concern related to elevated temperature and pressure

Within solid-state sintering regeneration, spent cathode materials are firstly mixed with Li salts, and subsequently, they are treated through different temperature levels in ranges of 600–900 °C. The stoichiometric ratio of Li is reinstalled by adding a supplementary Li source, and the particle bonding is indurated with high-temperature treatment [30,31].

Electrochemical relithiation is typically mediated in a three-electrode station; a spent cathode electrode stands as the working electrode, a platinum plate as the anode electrode, and one Ag/AgCl as the reference electrode with a Li-based solution. Optimal regeneration efficacy can be attained by tuning the concentration of the Li solution and changing the cathodic current. Nevertheless, this method cannot completely restore the cathode crystal structure, and additional temperature treatment is needed [7,32].

Ionothermal relithiation is a form of synthesis where ionic liquid solutions are simultaneously used as both the solvent and potential template or structure-directing agent in forming solids. This approach from the solution-based relithiation field evolves defect-targeted healing and homogeneous Li distribution inside cathode particles [33].

Eutectic molten salts establish a homogeneous environment during re-crystallization and particle growth stages of direct recovery at a eutectic temperature at ambient pressure. Initially, spent cathode materials are blended with Li-based eutectic molten salts; then, the resulting mixture is heated to the eutectic melting point, where re-lithiation and crystal rearrangement occur. Contrary to the previous methods, this approach reduces both the

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holding time and annealing temperature without compromising the restored material's crystallinity [7,34].

The selection of the optimal relithiation process depends on several crucial factors. These include not only the chemical composition of the active material but also the intensity and nature of defects present in the cathode structures, desired recovery efficiency, along with evaluations of investment and operating costs.

3. Limitation of Implemented Direct Recycling Procedures

Recycling of LIBs stands as a high-perspective strategy of their waste management, which addresses environmental concerns, marking a crucial step toward the sustainable development of energy storage technologies and the circular economy. In comparison to conventional hydrometallurgical and pyrometallurgical recycling methods, direct recycling serves several benefits in terms of process simplicity, low energy consumption, reduced secondary environmental pollution, and potential higher cost return [1,20,24].

While direct recycling has shown initial success within laboratory-scale settings, recovering a diverse array of cathode and anode materials, its transition to the industrial sector remains impeded. The practical application of this technology encounters significant challenges, signaling the need for further advancements in the research area [20].

The dominant limitation of currently implemented technologies resides in the processing of high-quality CBM or anode materials (black mass form) derived from the LIBs recycling pre-treatment, as shown in Table 4. The demand for ensuring high quality and uniformity in the resulting product increases the complexity of the entire technology and costs, thereby substantially diminishing the number of industrial implementations [1].

Table 4. Overview of the main limitations of currently implemented direct recycling techniques [1].

Limitation	Description
Limited applicability to whole spent LIB systems	The current approaches predominantly target the cathode and anode materials, neglecting other components of spent LIBs; thus, the potential for achieving a closed-loop circular economy for LIBs is not fully harnessed.
Black mass processing	The standard black mass, comprising a mixture of several materials, introduces technical barriers to the direct implementation of direct recycling techniques.
Energy-intensive recycling pre-treatment	Before direct recycling, LIBs undergo recycling pre-treatment processes involving high-temperature treatments and chemical interventions, which result in high energy consumption, emissions, and intricacies in the process of wastewater treatment.

Considering the commonly implemented LIBs recycling pre-treatment processes, whose output products consist of a mixed material output based on cathode and anode materials, conductive agents, PVDF binder, and other residual impurities from a current collector, or battery casings (Al, Cu, Fe, etc.) from a single type or different battery chemistry mixture [6], they pose significant technical obstacles, especially from the perspective of material separation or process efficiency, hindering the immediate progression of the direct recycling method [22]. A comparison of currently treated materials within direct recycling and the future strategy of this method is presented in Figure 5 from a comprehensive review work by Xu et al. [7].

The direct recycling methods are generally individually customized for various material treatments. They are accompanied by a range of limitations influenced by process complexity, readiness, and research status [7,22]. Thus, the direct recovery strategies, limitations, advantages, and disadvantages for crystal structure rearrangements for different types of cathode and anode graphite materials are discussed in the following sections.

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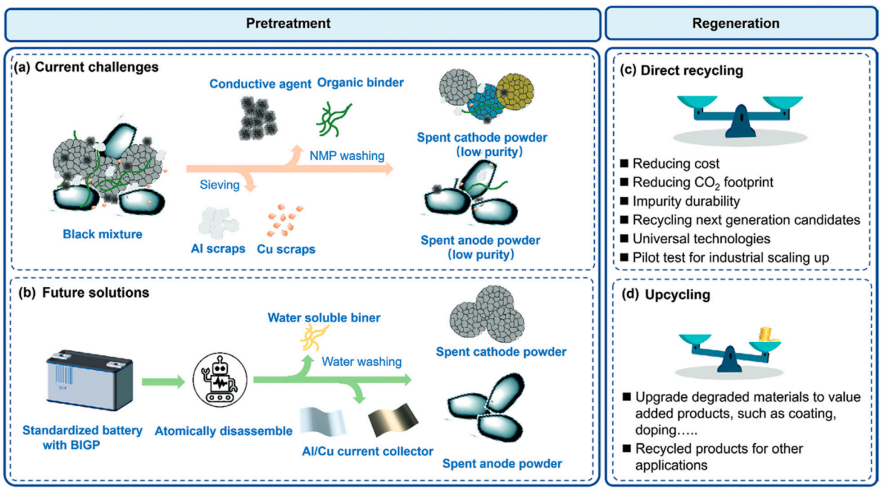

Figure 5. Current challenges (**a**) and future solutions (**b**) toward spent battery pre-treatment, as well as future development directions of electrode material regeneration (**c**) and upcycling (**d**) technologies. Reprinted from [7], with permission from Wiley-VCH GmbH, Copyright (2023).

3.1. Direct Recycling Treatment of Spent Cathode Materials

As a critical component of the battery, cathode materials play a crucial role in determining the overall performance, energy density, and efficiency of LIBs, where their differences within specific energy (capacity), specific power, safety, performance, lifespan, and cost are illustrated in Figure 6 [35].

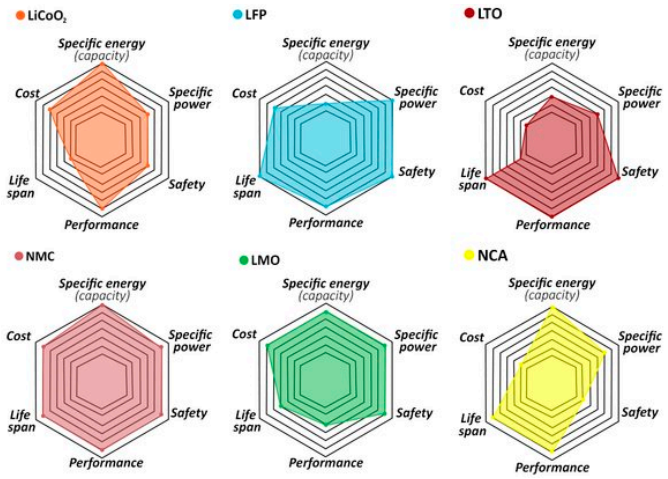

Figure 6. Comparison between different types of cathodes. Reprinted from [35] under the terms and conditions of the CC BY 4.0 license, Copyright (2021).

The demand for efficient energy storage systems has propelled research into enhancing the performance of LIBs and their suitable usage within selected application fields [36,37].

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Considering the current implementation stage and ratios between spent LIBs designed for EOL treatment, the spent NMC, LFP, LCO, and LMO cathode material treatments were described. More details about spent NCA materials' direct regeneration are provided, for example, in the study by Elmaataouy et al. [38].

3.1.1. Nickel-Manganese-Cobalt Oxide (NMC)

In the realm of electric vehicle (EV) applications, there is a distinct preference for battery technologies that offer higher energy density at reduced costs. Consequently, alternative ternary transition metal oxides incorporating Ni and Mn within LIBs, such as $LiNi_{0.33}Co_{0.33}Mn_{0.33}O_2$ (NCM111), $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ (NCM523), $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ (NCM622), and $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ (NCM811), have garnered widespread adoption and usage via different battery formats [6,39].

Spent battery cells of the NMC type commonly undergo severe phase transformations that result in the loss of Li-ions. The degraded cathode materials exhibit transformations involving spinel and rock salt phases on the surface of the particles. Thus, considering the relithiation techniques, short annealing is required within the treating processes to transfer spinel and rock phases back to a layered structure [7].

In the context of solid-state regeneration of NMC structures, layered cathode materials can be efficiently treated through standard sintering methods. Meng et al. [30] discovered that, before annealing NMC111 materials, milling plays a crucial role in enhancing the efficiency of electrochemical performance recovery by promoting uniform Li diffusion. The sintering properties vary based on the materials' constituents and crystallinity. As demonstrated by Shi et al. [40], polycrystalline NMC523 requires a sintering temperature within the range of 850 °C to 920 °C, while Gao et al. [41] recommend maintaining a temperature around 950 °C for single-crystal NCM523. For materials with a high Ni content, such as NMC622, Jin et al. [42] emphasize the necessity of a pre-oxidation treatment step before sintering. Additionally, higher Ni ratios demand increased oxygen pressure levels for effective treatment.

Within solution/hydrothermal-based relithiation, Shi's group [40] conducted comprehensive research on NMC523 recovery, achieving a recovered cathode material quality comparable to pristine conditions. The process involved a high-temperature short annealing (HT-SA) technique designed to suppress Li/Ni mixing and executed under specific oxygen atmosphere levels, as described in detail in the work by Gao et al. [41]. The reinstalled materials are subjected to minimal temperature and pressure loads, capped at 220 °C and 25 bar. Ongoing optimization efforts, such as those by Yu et al. [43], involve the introduction of redox mediation in the hydrothermal Li-bearing solution, resulting in a temperature decrease to 100 °C. In the context of NMC622 material regeneration, Sloop et al. [44] favored treatment with LiOH solutions. However, the efficiency encountered a setback due to the formation of a Li^+ and Ni^{2+} mixture.

The eutectic-mixture-based regeneration method for NMC materials has shown promising results. Shi et al. [34] utilized a eutectic solution consisting of lithium hydroxide (LiOH) and lithium nitrate ($LiNO_3$) to relithiate spent NCM523, successfully reverting the surface salt phase to its original layered structure, comparable to its pristine state. Fan et al. [45] also demonstrated effective regeneration of spent NCM523 using the LiOH and lithium carbonate (Li_2CO_3) system, which forms a molten salt. However, it has been observed that for highly degraded materials, this relithiation may not be optimal and might not fully recover destructed materials. New approaches are being explored, such as the solution proposed by Ma et al. [46], which involves adding cobalt(III) oxide (Co_2O_3) and manganese dioxide (MnO_2) to the eutectic solution to compensate for transition metal loss in cathode materials.

The research on ionic liquids for recovery faces significant limitations, primarily due to high implementation costs. However, as demonstrated by the results of Parnham et al. [47], the method can be effective and lead to full recovery, as shown in the case of NMC111,

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where an ionic liquid was served as the flux solvent, accompanied by LiBr as the Li source and ionothermal treatment.

The direct recycling technique for spent NMC materials can be considered an effective method for pristine-like cathode material recovery, with the possibility of using standard sintering methods and a lower environmental impact. However, it serves some notable limitations according to the applied relithiation technique, summarized in Table 5.

Table 5. Limitations and challenges of implemented direct recycling techniques focus on recycling spent cathode (NMC, LFP, LCO, and LMO) and anode graphite materials. "Pre-development phase" refers to an area that has not yet been thoroughly characterized by research. "Development phase" denotes a pilot research area with insufficient readiness for practical industrial application.

Spent Material	Solid-State Relithiation	Electrochemical Relithiation	Ionic Liquids Relithiation	Eutectic Salt Relithiation	Solution Based Relithiation
NMC	Necessity of milling before sintering; Long sintering time at high temperature; Pre-oxidation treatment procedures; Process complexity	Pre-development phase	High implementation costs	Destructed materials may not be fully recovered; Development phase	Formation of a Li^+ and Ni^{2+} mixture; HT-SA technique; Needs for oxygen atmosphere levels
LFP	High temperature treatment; Requisite of carbon and PVDF binders' removal from LFP particles; Structural stability; Sensitivity to sintering atmosphere	Development phase; Integration of pre-lithiated graphite; Poor industrial scalability	Pre-development phase	Development phase	Process complexity
LCO	Necessity of optimal Li/Co ratio; High energy process consumption	Process complexity; Requirement for high-quality delamination and purification steps	Pre-development phase	Development phase	Undescribed: thermodynamics and kinetics of implemented processes
LMO	High environmental demand; Best results: balanced molar ratio of LiOH and LMO, dependent on SOH	Pre-development phase	Pre-development phase	Pre-development phase	Necessity of optimal LiOH solution concentration; Not dependent on SOH
Graphite	High temperature treatment; Combined process (annealing, chemical leaching)	Pre-development phase	Pre-development phase	Pre-development phase	Necessity of impurities removal; Sensitivity to process atmosphere; High demand for wastewater treatment

3.1.2. Lithium Iron Phosphate (LFP)

Due to its good thermal stability, cost-effectiveness, and long cycle life, LFP technology plays a pivotal role in the LIB market, particularly in applications like portable electronics, grid energy storage systems, and EVs [48]. The absence of valuable metals, such as Co and Ni, in LFP cells reduces the economic motivation of their EOL treatment via conventional recycling methods [1]. Thus, implementing direct recycling methods with reduced energy-consumptive processes has the potential for wide-range extension in waste management [7].

The primary cause of capacity decay in LFP cells is the loss of Li inventory within the crystal structure while maintaining material stoichiometry. The regeneration process involves supplementing Li ions into the crystal structure. Familiar Li sources include inorganic Li, such as Li_2CO_3 or LiOH, lithium halides like lithium iodide (LiI) or lithium bromide (LiBr), and organic lithium salts, such as polycyclic aryl-lithium compounds [7].

The solid-state regeneration of LFP involves reloading the lost Li^+ ions in the cathode during cycling by annealing a mixture of Li-containing salt and spent cathodes at high temperatures between 350 and 700 °C [49]. However, the complete removal of conductive carbon and PVDF binder from LFP particles while preserving structural stability poses a challenge, affecting the quality of defect restoration and material purity. Research conducted by Li et al. [50] and Sun et al. [51] has demonstrated that the regenerated product is highly sensitive to the sintering atmosphere, where the inert or reductive atmosphere is preferred to prevent LFP oxidation, which can impact the final product quality.

Current research on electrochemical regeneration indicates that the electrochemical properties of the regenerated LFP are comparable to those of commercial materials. For

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instance, the study by Peng et al. [52] demonstrated that the recovered structure exhibited a discharge capacity of 135.2 mAh g^{-1} at 1 C, with a capacity retention of 95.3% after 500 cycles. However, the processes involve integration with a pre-lithiated graphite anode or separator, which may impose limitations on the industrial scalability of the process.

The solution-based recovery method for LFP materials holds promise for restoring both Li losses and the formation of Fe-Li defects, particularly in cases of severe degradation and substantial capacity decay [53]. The revival of these defects is crucial for achieving complete material recovery and optimal performance. However, the complexity of this method is a limitation, as demonstrated in the work by Panpan et al. [54], where a procedure involving strong electrostatic repulsion with various reduction agents was employed in a LiOH solution.

The main benefit of eutectic solution regeneration is that it covers relatively low-temperature treatment primarily via a reductive atmosphere, which increases the possibility of an LFP structure oxidation decrease [7]. Nevertheless, the crucial results of this method are under investigation in ongoing research.

The LFP direct regeneration covers several benefits, such as the recovery of high-quality materials with comparable electrochemical properties to commercial materials; nevertheless, the procedure covers some of its limitations, which are shown in Table 5.

3.1.3. Lithium Cobalt Oxide (LCO)

Batteries based on LCO structures find extensive applications, especially within the portable electronic field, due to their high energy density and compact size [6]. A notable reduction in Li content within the bulk crystal structures occurs throughout the degradation process, accompanied by significant damage to the surface of spinel phases. Consequently, recovery efforts focus on replenishing Li deficiencies and restoring degraded spinel phases to their original layered form. The relithiation process typically initiates in a solid or aqueous medium and progresses through annealing treatments [7].

Solid-state regeneration of LCO involves directly mixing degraded cathode materials with solid lithium sources, such as Li_2CO_3 or LiOH, followed by thermal treatment within the range of 800–950 °C. According to findings by Gao et al. [55] and Chen et al. [56], successful repair of material composition and structural defects occurs when the Li/Co ratio falls within the range of 1.0–1.2. A lower ratio results in incomplete structural repair, while a higher ratio introduces Li_2O impurities, negatively impacting the final performance.

The electrochemical relithiation approach for LCO materials encompasses several highly efficient procedures, with the primary drawbacks residing in process complexity and higher cost. Yang et al. [57] outlined a regeneration technique for cycled LCO in their work. The spent cathode material (working electrode) was paired with a new LCO electrode, the electron donor, and the Ag/AgCl reference electrode. During discharge, ions from the Li_2SO_4 electrolyte infiltrate damaged structures, reinstating Li deficiencies. A notable advantage was the absence of observed Al corrosion on the working electrode. After a brief annealing process, the cathode structure fully recovered, exhibiting capacity and cycle performance on par with the pristine commercial batch. However, a key limitation of this method is the necessity for high-quality delamination and purification steps in regenerating materials.

The solution-based regeneration of spent LCO requires further exploration, particularly regarding microstructure revitalization and the Li^+ replenishment process in an aqueous solution. Current results, as presented in the work by Kim et al. [58], describe a regeneration conducted at 200 °C for 20 h in a LiOH solution. The resulting structure of LCO demonstrated a capacity of 144 mAh g^{-1} with 92.2% retention after 40 cycles. Additionally, Shi et al. [59] conducted experiments involving highly effective hydrothermal relithiation, followed by post-sintering; however, the thermodynamics and kinetics of these steps remain uncharacterized.

The eutectic solvent regeneration offers several advantages, including non-toxicity, low cost, and recyclability. However, these procedures are primarily in the development

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phase. As an illustration, Wang et al. [60] present a well-designed procedure using the lithium chloride (LiCl) and urea (CH_4N_2O) eutectic system, which features a reduced eutectic point to below 120 °C, for the direct regeneration of degraded LCO.

Compared to other recovery techniques, direct recycling of spent LCO materials serves several advantages, such as low environmental impact and the absence of Al corrosion. However, they are connected to the limitations summarized in Table 5.

3.1.4. Lithium Manganese Oxide (LMO)

The prominence of LIBs with Mn-rich cathodes is increasing in EVs and large-scale energy storage due to their stable performance and cost-effectiveness [7]. This trend is particularly notable in scenarios where high energy density is not the primary consideration. Likewise, for LFP recycling, the economic viability of traditional pyrometallurgical and hydrometallurgical recycling methods for recovering Mn compounds from spent LMO cathode materials is constrained by their relatively low market value [24]. Consequently, the economic motivation for the recycling of LMO is circumscribed. In this context, direct recycling emerges as a pertinent and economically viable strategy for effectively managing spent LMO cathode materials [5].

The deterioration of LMO materials is due to Mn^{2+} dissolution [61], resulting from the disproportionation of Mn^{3+} [62]. In the initial charge, Mn^{3+} undergoes a reaction, forming spinel-type $Li_4Mn_5O_{12}$ and soluble trimanganese tetraoxide (Mn_3O_4). During discharge, $Li_2Mn_2O_4$ spinels are generated with Jahn–Teller distortion. As a benefit, cycled LMO retains the spent spinel LMO phase without impurities [7].

Regarding the recovery of spent LMO materials, solid-state and electrochemical (aqueous) relithiation regeneration are the preferred methods. Typically, LiOH solutions in various concentrations are utilized to treat structural defects. However, as demonstrated in review work by Panpan et al. [7], highly concentrated solutions exceeding 0.4 M induce the formation of Li-rich phase lithium manganite (Li_2MnO_3) impurities during the recovery process. Wang et al. [63] have shown that hydrothermal methods yield superior recovery results compared to solid-state sintering in terms of phase purity and electrochemical capacity of the treated LMO. Solid-state techniques restore the lost capacity by maintaining a well-balanced molar ratio of LiOH and aged LMO, dependent on the degraded materials' state of health (SOH). In contrast, hydrothermally recycled products recover lost capacity irrespective of the SOH of the spent LMO. The primary difference among these approaches is reflected in their energy and environmental implications, with hydrothermal processes demonstrating markedly reduced energy consumption compared to solid-state sintering, as provided in the LCA study by Xio et al. [64].

Although the direct recycling of spent LCO cathodes offers low energy consumption, it also has some limitations, as detailed in Table 5.

3.2. Direct Recycling Treatment of Spent Anode Materials

Graphite stands out as a frequently used anode material in LIBs [65], attributed to its excellent electrical conductivity, high thermal and mechanical stability, prolonged cycle life, good capacity, and low working voltage. Nevertheless, the recycling of spent graphite anodes received comparatively less focus due to the absence of transition metals. Thus, spent anodes are either incinerated or disposed of in landfills. Considering the potential demand for graphite minerals, which is projected to be approximately 13 times higher than the demand for Li minerals, especially in the context of the 2025 EV market, there is a critical need to streamline and enhance the recycling processes for these materials [66].

Several research groups have devoted their work to graphite direct regeneration [67,68]. Yang et al. [69] introduced a comprehensive direct recycling perspective for all anode material recovery, including graphite, Cu foil, aluminium hydroxide ($Al(OH)_3$), and Li_2CO_3. This approach combines heating and leaching treatment steps and leads to the recovery of 99.9% pure graphite with significantly improved specific capacity and excellent cycling performance. The anode materials may contain impurities such as transition metals, which,

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as demonstrated by Ma et al. [70], can be effectively treated and removed through sulfuric acid (H_2SO_4) and hydrogen peroxide (H_2O_2) leaching. However, this approach increases the complexity of wastewater treatment, posing a limitation to full-scale industrial applications. In response, Wu's [71] and Wang's [72] research groups focused on cleaner methods, leveraging the highly reactive properties of LiC_x in spent anode materials with water, enabling the removal of residual Li from graphite particles. The oxidation degradation of LiC_x is mitigated by an inert atmosphere, albeit resulting in higher costs.

Acid or water treatments prove effective in eliminating impurities from spent graphite anodes. However, subsequent high-temperature annealing steps become imperative to attain the complete recovery of graphite anodes. A review by Panpan et al. [7] reveals atom rearrangement and structural recovery after a 6 h treatment at 3000 °C in an N_2 environment. Nevertheless, the primary limitation lies in their high energy consumption.

The EOL processing of spent anode graphite-based materials is currently in the developmental stage and requires further optimization, as current approaches indicate. However, the limitations of the implemented direct recycling methods for treating spent graphite anode materials are outlined in Table 5.

4. Discussion

Recycling spent LIBs represents an effective waste-management strategy that mitigates the environmental impacts by preserving raw resources, especially valuable metals, reducing the emission footprint of mining and post-processing, and fostering the principles of a circular economy. Given the prevalent global practices in recycling, such as pyrometallurgy or hydrometallurgy, which often entail high energy consumption, emissions, or the extensive use of chemicals, a pressing need arises for alternative recycling technologies. These alternatives should enable the direct reutilization of deteriorated energy storage materials in battery manufacturing, aligning with economic and environmental sustainability objectives.

The currently employed direct recycling methods focus on treating high-quality cathode or anode materials obtained through the recycling pre-treatment of the same type or different types of LIBs. Consequently, these direct regeneration approaches do not encompass the treatment of entire spent LIB systems; instead, they specifically target the most economically valuable products in the EOL treatment chain, commonly referred to as the black mass. During the recycling pre-treatment phase, materials are subjected to high-temperature treatments, incurring substantial operating costs and emissions. Moreover, these materials may undergo treatment with various chemicals, introducing complexities to wastewater treatment or necessitating specialized equipment to ensure compliance with safety regulations and produce high-quality end products. The combined approach of pre-recycling and the selective recycling of specific materials, as illustrated in Figure 7, significantly influences the operation's environmental and economic aspects, impacting the overall motivation for the industrial implementation of direct recycling techniques.

Direct recycling methods are typically customized for specific material treatments. Presently, "relithiation" techniques, encompassing solid-state sintering, electrochemical relithiation in organic and aqueous electrolytes, ionothermal relithiation, solution relithiation, and eutectic relithiation, are predominantly utilized for the regeneration of cathode materials. This review provides an overview of the direct recycling of LIB cathode types, including NMC, LFP, LCO, and LMO, and graphite anodes generally in laboratory scaled-up treatments. Nevertheless, significant challenges persist in applying these techniques on a larger industrial scale.

While individual methods exhibit high efficiency in restoring the properties of regenerated materials, bringing them to a comparable level with pristine ones, the primary challenge in direct recycling lies in processing black mass obtained from different LIB types. The product typically consists of shredded cathode and anode materials, conductive agents, PVDF binder, and various residual impurities (Al, Cu, Fe, etc.). Material diversity sets several technical burdens for subsequent processing, such as product separation or

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implementation complexity. These additional treatments represent a vital process stage in the current research state. They aim to achieve highly efficient direct recycling results while increasing economic demands and environmental burdens.

Figure 7. Current direct recycling process, including recycling pre-treatment and recycling steps, covering application of different relithiation techniques.

Furthermore, it is essential to consider the potential application field of regenerated materials. When considering currently common optimizations, covering alterations in material ratios or substitutions of utilized compounds, it is crucial to assess the potential of complex regeneration processes compared to the regeneration of chemical compounds individually. The introduction of direct recycling processes should be up-to-date and fully respect the demand of the battery market. If the current development trend continues to be maintained, regenerating outdated battery structures would not be an advantage and, on the contrary, could bring significant economic losses.

Before full-scale industrial implementation, careful consideration of the mentioned factors is imperative. Presently, direct recycling remains in its beginning stages. Research predominantly revolves around simplified models, focusing on single-type materials or black mass sourced from specific battery types. Most processing occurs at a laboratory scale, with pilot operations representing the extent of practical application thus far. Only through more thorough evaluations can it be determined whether direct recycling will emerge as a distinct, standalone recycling approach for managing waste LIBs or whether it will function as an adjunct process alongside established techniques.

An overview of the limitations of selected direct recycling techniques identified within this work, depending on the processed material, is shown in Table 5. Besides these summarized limitations for common cathode and anode materials, state-of-the-art direct regeneration technologies need more exploration and optimization within the following areas, which are discussed point-to-point subsequently.

- Direct recycling pre-treatment;
- Complex material composition;
- Black mass processing;
- Environmental and economic impact;
- Perspective of industrial scale.

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4.1. Direct Recycling Pre-Treatment

Current direct recycling methods mainly focus on high-quality cathode or anode materials (black mass) derived from recycling pre-treatment steps, which ensure the removal of unsolicited materials. Thus, integrating recycling pre-treatment into the EOL process is essential; however, it leads to the non-recovery of selected material groups and significant process losses. Furthermore, the procedures become increasingly technologically complex when considering the simultaneous processing of various chemical types of batteries, a scenario that is quite common across other recycling techniques. The pre-treatment commonly consists of complex chemical treatment followed by energy- and emission-demanding steps, which decrease the environmental and economic benefits of direct recycling procedures. Therefore, future optimization should involve comprehensive treatments suitable for the entire battery system, ensuring a closed-loop circular economy for LIBs, and maximizing material recovery and process efficiency.

4.2. Complex Material Composition

Current methods commonly deal with the processing of single-type materials or the processing of cathodes and anodes of one type of battery. However, conventional recycling often involves processing various battery types, producing multi-material black mass. If it is not technologically possible to process this type of product via direct recycling, it would be necessary to extend the recycling pre-preparation by, e.g., a sorting step, where the batteries would be classified according to their chemistry type. In this context, leveraging the Battery Passport system would be feasible and logistically appropriate. Otherwise, processing and sorting the black mass would be necessary by implementing proper separation techniques. Moreover, it is imperative to consider the advantages of recovering entire cathode structures, while also accounting for technological advancements and preferences within changes in material ratios. This includes exploring optimal methods for industrial-scale recycling, considering the aging of technology and new possibilities for their utilization.

4.3. Black Mass Processing

Although high-quality black mass can be obtained as a typical product of recycling pre-treatment, there is a high possibility that it will contain impurities in the form of residual current collectors, organic materials, system casings, or anode/cathode materials in the scenario of direct processing of spent cathodes/anodes. Thus, before the direct recycling procedure itself, it is necessary to add a series of additional treatment steps, such as high-temperature annealing, chemical leaching, or precipitation post-treating, to remove impurities and achieve the desired high efficiency of the process. Nevertheless, this sequence of procedures is characterized by high energy consumption, significant emissions, and the generation of additional waste products, all of which contribute to elevated process costs and a larger overall environmental footprint. Thus, future optimization efforts should still be focused on developing effective methods to separate and treat black mass, ensuring the recovery of valuable materials, minimizing impurities, and achieving as high a purity level in regenerated materials as possible. Nevertheless, the processing and purification of black mass present a drawback inherent in currently implemented procedures, mainly for the pre-recycling lines, with the output product constituting black mass, whose quality directly influences its market value. Mitigating energy consumption and minimizing the environmental impact of these procedures would benefit all waste LIB recycling processes.

4.4. Environmental and Economic Impact

Direct recycling methods should be optimized to minimize energy consumption, economic demands, and environmental impact. Current direct recycling processes involve high-temperature treatments, such as annealing, smelting, or roasting, and chemical interventions, leading to elevated energy consumption, emissions, and complications in wastewater treatment, mainly as part of recycling pre-treatment or additional separation

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procedures as part of black mass post-processing. Future optimization efforts should prioritize environmentally sustainable practices and ensure an eco-friendlier approach. The incorporation of environmentally friendly processes has the potential not only to contribute to sustainability but also to impact operational costs positively over subsequent years. By adopting eco-friendly practices, companies can anticipate reducing the required investment, for example, through specialized equipment prices. This dual benefit—environmental sustainability and cost effectiveness—creates a favorable environment for the advancement and widespread adoption of these processes.

4.5. Perspective of Industrial Scale

At present, direct recycling is primarily encountered at the laboratory level. The introduction of effective procedures on this smaller-scale level represents an initial step that can ensure fewer demanding processes from environmental and economic perspectives when fully implemented on the industrial level. To ensure effective EOL management, especially of LIBs, it will be necessary to evaluate the possibilities offered by direct recycling and to focus on pilot projects and recycling plants, where processing at the industrial scale level is presented. Based on that, decisions can be made regarding whether direct recycling will be replaced or implemented as a supplemental procedure to the currently established methods.

Considering current results and the wide range of optimization possibilities, there is a high potential for global and large-scale implementation of direct recycling as the major or supplementary EOL treatment technique for spent LIBs. Moreover, it can be perceived as a sustainable and eco-friendly alternative to conventional recycling processes because it complies with crucial environmental, economic, and technological factors. By minimizing the use of harsh chemicals and reducing energy consumption and emissions of high-temperature treatments, these methods contribute to a smaller carbon footprint and align with global efforts toward environmental sustainability. Regarding the increase in environmental legislative restrictions, LIB's waste management requests, and market trends that prioritize sustainability, the demand for eco-friendly battery recycling methods grows. Thus, direct recycling can be positioned as a key player in addressing these evolving dynamics and fostering a more sustainable future. Nevertheless, the significant shortcomings of current direct recycling processes must be considered and addressed before full-scale implementation.

5. Conclusions

This review outlines the limitations of current direct recycling methods for common cathode materials (NMC, LFP, LCO, and LMO) and anode graphite in spent LIBs, mainly from a laboratory-scale perspective. The work points out the main limitations of state-of-the-art direct recycling technologies, including solid-state sintering, electrochemical relithiation, ionothermal relithiation, solution relithiation, and the eutectic relithiation method, which are typically used for highly effective regeneration of spent materials, including the restoration of their electrochemical and structural properties. Additionally, this work summarizes the main challenges of selected direct recycling methods, which are essential for potential optimization steps leading to larger-scale (fully industrial) applications. The current techniques of direct recycling typically treat the output of recycling pre-treatment, the black mass. The necessity of energy-demanding pre-treatment steps impairs the perspective of subsequent, direct regeneration techniques and is the dominant limitation of current industrial implementation. Thus, this review emphasizes the critical constraints and the need for further optimization within direct recycling, representing a promising EOL treatment for spent LIBs in future years.

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Review

Literature Review, Recycling of Lithium-Ion Batteries from Electric Vehicles, Part II: Environmental and Economic Perspective

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Abstract: Lithium-ion batteries (LIBs) are crucial for consumer electronics, complex energy storage systems, space applications, and the automotive industry. The increasing requirements for decarbonization and CO_2 emissions reduction affect the composition of new production. Thus, the entire automotive sector experiences its turning point; the production capacities of new internal combustion engine vehicles are limited, and the demand for electric vehicles (EVs) has continuously increased over the past years. The growing number of new EVs leads to an increasing amount of automotive waste, namely spent LIBs. Recycling appears to be the most suitable solution for lowering EV prices and reducing environmental impacts; however, it is still not a well-established process. This work is the second part of the review collection based on the performed literature survey, where more than 250 publications about "Recycling of Lithium-ion Batteries from Electric Vehicles" were divided into five sections: Recycling Processes, Battery Composition, Environmental Impact, Economic Evaluation, and Recycling and Rest. This paper reviews and summarizes 162 publications dedicated to recycling procedures and their environmental or economic perspective. Both reviews cover the techno-environmental economic impacts of recycling spent LIBs from EVs published until 2021.

Keywords: lithium-ion battery; electric vehicles; recycling processes; environmental impacts; economic evaluation; battery recycling; battery reuse; electric vehicles; literature review

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1. Introduction

Lithium-ion batteries (LIBs) represent the dominant energy source not just for portable electronics and electromobility vehicles, but they have also found growing popularity in military and aerospace applications or grid energy storage systems [1]. The ever-increasing demand for LIBs leads to the still-growing amount of battery waste, from the small formats of the battery cells to complex automotive or storage battery systems.

The entire automotive industry is undergoing a crucial turning point. The increasing requirements for decarbonization and CO_2 emissions reduction limit the production of new combustion vehicles [2], and transit to the production of electric vehicles (EVs) is the most suitable solution. Therefore, pressure to reduce EVs' price and a detailed evaluation of their environmental impacts are necessary. Thus, these systems' environmental and economic (E&E) assessments are inherent.

LIBs from EVs are commonly based on valuable metals, such as nickel (Ni), cobalt (Co), manganese (Mn), and lithium (Li), which are significantly high in cost. Packaging and coverings are created from an efficiently recyclable material such as aluminum (Al) or copper (Cu). Thus, one of the most suitable waste management solutions for spent LIBs seems to be recycling processing. Introducing recycling techniques will lead to the recovery of selected materials as high-quality outputs; thus, raw materials extraction, including high energy consumption and CO_2 emission production, could be reduced [3,4]. The price of

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a new battery based on reused materials should be lowered, as well as the price of the new EV. These days, the research is mainly dedicated to optimizing the currently operated recycling techniques, reducing their complexity, and increasing output efficiency and purity. Nevertheless, the environmental and economic demands of selected recycling routes are being examined and compared to each other; LIBs technology is evaluated within the entire life cycle or recycling framework's financial burdens.

This paper presents the second part of the literature review study of peer-reviewed publications devoted to "Recycling of Lithium-ion Batteries from Electric Vehicles", focusing on the E&E perspectives. In total, 162 papers have been referred to in this work and summarized into individual thematically connected units (categories), including, for example, Life Cycle Assessment (LCA) study, recycling and recovery of materials, or economic evaluation. In addition, conclusions for individual reviewed categories are provided, and recommendations for further research and development are proposed.

Both published reviews cover the entire techno-environmental economic impact of recycling EV-retired LIBs published until 2021. More than 90% of used publications from both reviews have been published in the last decade. Thus, the current overview that captures individual steps of the recycling techniques, their impacts on the environment, and the economic demands are presented. These works provide a comprehensive techno-environmental economic analysis of the selected field, and it is prepared for further use in academic and industrial spheres.

The content of this work is structured as follows: firstly, in Section 2, the methodology of the performed literature review is shortly discussed; then, in Section 3, the current state of performed recycling techniques, their recovery efficiencies primarily in terms of valuable metals considering their environmental impacts considering LCA study and economic assessments inclusive of financial analysis and evaluation are reported. Finally, in Section 4, the main conclusions of this part of the review collection are pointed out, and both parts are discussed comprehensively to capture the recycling of LIBs from EVs as a technological environmental economic unit.

2. Review Methodology

This paper is based on the literature review that has already been introduced in the first part of the review collection devoted to recycling technology of LIBs from EVs [5]. The performed search was completed during September and October 2021 and was performed on two databases, Web of Science (WoS) and Scopus, focusing on the variation of terms "recycling", "Lithium-ion", and "electric vehicles". A more detailed description of this search methodology and enumerations of individual publications is presented in the first part of the literature review study devoted to recycling technology [5].

This second part of the review summarized the remaining 250 publications, including articles, reviews, proceedings papers, early accesses, editorial materials, and corrections dedicated to the environmental and economic (E&E) approach. As the results of the first part of this review collection show, there is an overlap in many scopes of selected publications. This overlay is presented in the updated Table 1. Moreover, it illustrates the distribution of chosen publications into five sections (Recycling Processes, Battery Composition, Environmental Impact, Economic Evaluation, and Recycling and Rest), which are being represented by a specific category/keyword according to the content.

In total, 162 publications were one-by-one reviewed in this paper from the environmental economic field to which this work is dedicated. Thus, this paper primarily focuses on the Battery Composition, Environmental Impacts, Economical Assessment, and Recycling and Rest (R&R) sections and their relevant categories. Moreover, Appendix A provides a brief overview, including the type, publication year, and summary content of all reviewed publications.

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Table 1. Distribution of publications; the methodology of performed literature review.

Section	No. of Articles in Section	Category/Keyword	No. of Articles in Category	No. of Overlapped Articles
Recycling Processes	61	Pretreatment	3	0
		Metallurgy/Mechanical	6	1
		Pyrometallurgy	4	0
		Hydrometallurgy	19	11
		Direct Recycling	6	3
		Special Method	23	9
Battery Composition	54	Cathode	43	43
		Anode	9	9
		Electrolyte	2	2
Environmental Impact	75	Envi/Ecological Impact	13	5
		Life Cycle Assessment (LCA)	5	3
		Recovery of Materials	35	1
		Recycling of Materials	22	0
Economical Assessment	14	Economical Evaluation	14	3
Recycling and Rest (R&R)	47	Recycling EV LIBs	11	6
		Recycling LIBs	36	19

3. Results

Within this part of the review collection, the categories of Battery Composition, Environmental Impact, Economical Assessment, and R&R were discussed. As in the previous work, the representation of individual types of publications in these distinctive sections was evaluated. A total of 124 Articles, 17 Reviews, 18 Proceedings Papers, 2 Editorial Materials, and 1 Correction were used to characterize individual recycling procedures and E&E aspects. The representation of the publications' distribution in categories is shown in Figure 1.

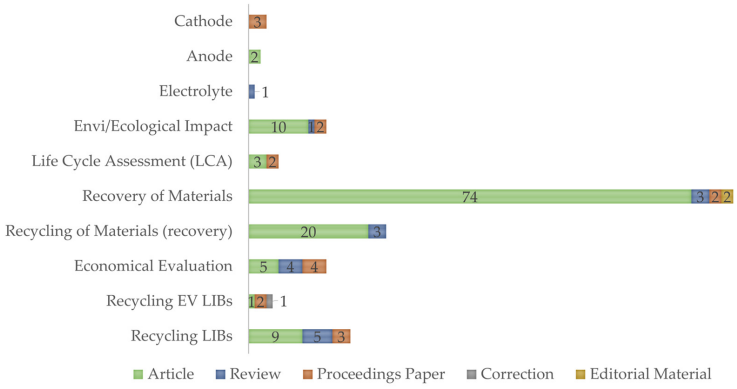

Figure 1. Distribution of reviewed publications according to their type.

The recycling process itself stands as a very complex topic. Thus, the distribution of the publication into individual categories is not fully restrictive and unambiguous. The reviewed publications were sorted and organized for the most straightforward interpretation of recycling LIBs from EVs under the environmental and economic perspectives. The methodology of the work is presented in Figure 2.

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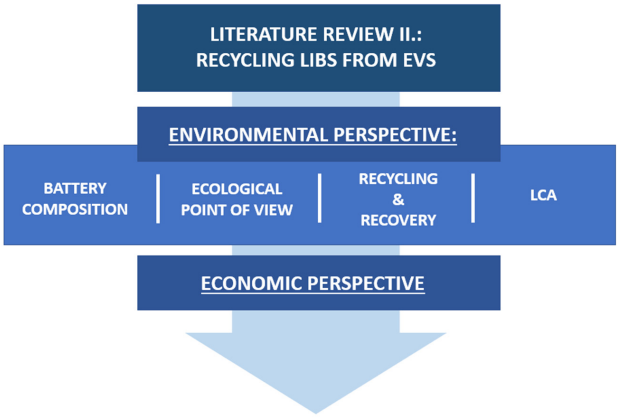

Figure 2. Methodology of the work devoted to E&E of the recycling LIBs from EVs.

3.1. Environmental Perspective

The introduction and application of technology are not assessed only in terms of its effectiveness, efficiency, or economic feasibility of implementation. A significant role stands the impact on the environment, monitored within the individual phases and primarily during the entire life cycle.

LIBs have been developed since the last decade of the 20th century. The quality of the technology used and its application level are growing year by year, whether in terms of small portable devices (mobile phones, laptops, etc.) or electromobility (e-bicycles, e-scooters, EVs). Due to the ever-tightening rules of the EU Directive 2019/631, which proposes a reduction of CO_2 emissions of passenger vehicles and vans [2], a more significant increase in demand for EVs could occur. It could lead to a 45% growth of EVs in the total market by 2030 [6].

As EV production increases, the number of retired LIBs from these cars increases too. Thus, addressing environmental impact issues is progressively essential. Primarily, it is advisable to focus on the new battery production (used technologies and materials), effective use during an active life, and evaluate the efficiency and burden of the disposal or recycling process.

According to this work, the publications evaluating the environmental point of view were divided into four categories.

- Battery Cell Composition, which deals with the structure of the cells.
- Ecological point of view that addresses a purely ecological point of view.
- Recycling Processes and Recovery Efficiencies, which provides a detailed summary of applied recycling techniques, their efficiencies, and findings.
- LCA (Life Cycle Assessment) study that evaluates the life cycle of the battery.

An overview of frequently used abbreviations of main indicators in the framework of environmental studies or impacts monitoring, which are used later in the text, is provided in Table 2.

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Table 2. Overview of the main indicators for monitoring the environmental impact.

Abbreviations	Indicator
ADP	Abiotic resource depletion potential
CED	Cumulative energy demand
GHG	Greenhouse gas
GWP	Global warming potential
TETP	Terrestrial ecotoxicity potential

3.1.1. Battery Cell Composition

As is generally well known, there are four main components of a battery cell: two terminals (electrodes) based on different chemicals (typically metals), the anode and the cathode, the electrolyte, and a separator. The electrolyte is a chemical medium that ensures the flow of electrical charge between the electrodes. Furthermore, the battery cell contains current collectors, typically formed by metals such as Cu or Al.

LIBs contain roughly 5–20 wt.% cobalt (Co), 5–10 wt.% nickel (Ni), 5–7 wt.% lithium (Li), 5–10 wt.% other metals (manganese (Mn), copper (Cu), aluminum (Al), iron (Fe), graphite (C), etc.), 15 wt.% organic compounds, and 7 wt.% plastics. The proportion varies according to exact cell composition and the manufacturer [7,8]. The general main material composition of the individual components of LIBs—anode, cathode, and electrolyte, which is typically dissolved in propylene carbonate, ethylene carbonate, or dimethyl sulfoxide—is shown in Table 3. Binders, flame retardants, gel precursors, and electrolyte solvents complement these components [8–10].

Table 3. The general material composition of the individual components of LIBs [8–10].

Anode	Cathode (Short Name)	Electrolyte
Metallic lithium	Lithium manganese oxide (LMO)	$LiPF_6$
Graphitic carbon	Lithium cobalt oxide (LCO)	$LiClO_4$
Hard carbon	Lithium nickel cobalt manganese oxide (NMC)	$LiAsF_6$
Synthetic graphite	Lithium iron phosphate (LFP)	$LiCF_3SO_3$
Lithium titanate	Lithium nickel cobalt aluminum oxide (NCA)	$LiBF_4$
Tin-based alloys	FeS_2	$LiSFO_2$
Silicon-based materials	V_2O_5	
	Electronic conducting polymers	

According to the performed search, the topic of "Battery Cell Composition" has not been intensively focused on by researchers since it was only addressed in six publications, including three Proceedings Papers, two Articles, and one Review, directly dedicated to the individual components of the retired LIBs from EVs.

A detailed and very well-structured review, dedicated to the recycling with emphasis on the anode (graphite) and electrolytes, was prepared by Arshad et al. [11]. This work presents essential elements of Life Cycle Assessment (LCA), focusing on End-of-Life (EOL) LIBs recycling, process limitations, and future efforts to improve the current efficiency of metal extraction and separation.

In compliance with this work, the anode and electrolyte are typically decomposed by heating (pyrometallurgy) or filtration/distillation (hydrometallurgy). Calcination or pyrolysis is habitually used for the high recovery rate of anode materials, as well as for electrolytes [11]. The mechanisms of these techniques were presented in the first part of this literature review collection [5], or it is described in more detail in the full version of the referred paper [11]. In addition, the work provides a summary of potential applications of recycled materials. The recovered carbon-based anode materials can be used as absorbents, nanocomposite thin film (in conjunction with polymers), or as part of a new anode in LIBs. Recycling and reusing electrolytes are not efficient under current conditions due to their fast capacity fade during repeated use [11].

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The procedures, recovery rates, or a future application potential of recycled cathode materials have not been described in the summary publication yet. Selected publications deal with specialized implemented recycling procedures. Wen et al. [12] and Zhou et al. [13] discussed basic recycling procedures (pyrometallurgy, hydrometallurgy) supplemented by biological recycling; Meng et al. [14] provided a recycling strategy for spent graphite/$LiFePO_4$ batteries and complemented a cathode composition based on recovered LFP/graphite with cation/anion-co-storage capability, which design leads to the new-type dual-ion battery. Sloop et al. [15] presented the advantages of direct recycling of electrode materials and compared the process for bare and coated NMC 622 cells. Wu et al. [16] devoted their study to the physical separation process, implemented via thermal and mechanical treatments that recover active cathode materials ($LiFePO_4$) from current collectors (including Al fragments); the process is based on cohesive zone models verified by physical separation experiments.

3.1.2. Ecological Point of View

According to the chemical composition of batteries, they are dominantly based on precious metals such as Co, Ni, Li, Mn, and other frequently used metals, e.g., Cu and Al. The secondary use and recovery of these metals as high-quality recycling products bring many advantages, where the natural resource savings resonate the most.

Current and future environmental impacts of recycling in close-loop battery life are being addressed in many cases. It leads to the introduction and ever-tightening conditions of legislation, for example, the currently valid EU legislation (increasing recycling efficiency from 50% to 65% by 2025 and 70% by 2030 by average weight [17]) or growing efficiency for the constitution of current recycling methods. They are discussed in detail in 13 publications, i.e., in 10 Articles, 2 Proceedings papers, and 1 Review, which majorly deal with the ecological principles.

These sorts of questions are discussed in the critical analysis of natural resource savings published by Dewulf et al. [18]. This paper compares a detailed analysis of a LIB recycling scenario, including Co and Ni recovery and re-introduction into the battery production chain, with a virgin production scenario. The (a) input exergy that is necessary and allocated for the production chain of 1 kg LIB cathode material, where Scenario A describes the production of a battery based on waste with Ni and Co recovery, and Scenario B shows production from virgin resources; and (b) the contribution to the total natural resource consumption as a function of the involved processes, that includes their supply chains, are shown in Figure 3. The exergy and cumulative exergy extracted from natural sources are used for savings quantification. According to the results, the recycling scenario leads to a 51.3% natural resource savings, caused primarily by decreasing mineral ore dependency and reducing fossil resources (45.3% reduction) and nuclear energy demand (57.2%). Moreover, the paper discusses the economic benefit of recycling.

Rahman et al. further investigated the impact of reusing recovery materials during a new LIBs production [19]. This research was devoted to recovering active materials of a spent 48.8 Wh LIB ($LiCoO_2$) by applying the laboratory-scale hydrometallurgical procedure. The result shows that the recovery of active metals is 41% of the cathode and 8.5% of the anode materials. Due to this process, the generated emissions can be reduced by 47.61% in the case of metal production for batteries and 60.7% for disposal transportation of the spent battery. The emission can be reduced by 52.85% by recycling the active materials in this type of LIB.

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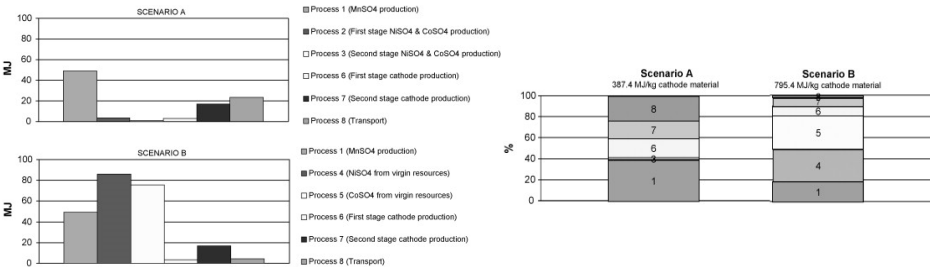

(**a**) (**b**)

Figure 3. (**a**) Inputs (exergy, MJ) necessary and allocated for the production chain of 1 kg Li-ion battery cathode material. Scenario A: starting from battery waste with Ni and Co recovery. Scenario B: from virgin resources; (**b**) Contribution (in%) to the total natural resource consumption as a function of the involved processes, including their supply chains. Reprinted from Resources, Conservation and Recycling, Vol. 54, Dewulf et al. [18], Recycling rechargeable lithium-ion batteries: Critical analysis of natural resource savings, no. 4, pp. 229–234, Copyright (2022), with permission from Elsevier.

The analysis presented by Dunn et al. [20] answers three critical questions about automotive LIBs energy and environmental impacts:

- Is it materials production or battery assembly that causes more of these impacts?
- What motivates battery recycling if it is the assembly step that is the primary energy consumer?
- How do the energy and environmental performance of EVs and internal combustion engine vehicles (ICVs) compare?

The study provides several additional questions and answers that confirm the obtained results, including, for example, local impacts of the recovery of metals in cathode materials' supply chains, including high SO_x emissions. Due to the complexity of the topic, the details and the obtained results can be found in the full version of this paper [20].

When comparing the effects of recycling, it is necessary to consider the mutual comparison of currently used recycling methods; this is where the publication by Costa et al. [21] is focused. The advantages and disadvantages of the different current-state techniques are discussed in their work; these are shown in Table 4. Moreover, the environmental issues associated with EV LIBs' production, use, and EOL procedures are considered and described.

Table 4. Comparison of used recycling techniques. Reprinted from Energy Storage Materials, Vol. 37, Costa et al. [21], Recycling and environmental issues of lithium-ion batteries: Advances, challenges and opportunities, pp. 433–465, Copyright (2022), with permission from Elsevier.

Method	Advantages	Disadvantages
Direct recycling	Environmentally friendly; High specificity; Non-destructive method.	Non-specific; Not possible processing of different cathode materials.
Pyrometallurgical method	High recycling rates; Solvent free.	High-temperature process; Other processes for the effective recovery of materials are necessary.
Hydrometallurgical method	High recycling rates; Large variety in the recovery of metals.	Complexity of the process; Application of toxic reagents; High in costs.

The environmental impacts are well overviewed in work by Martins et al. [22] and in a comprehensive review by Meshram et al. [23]. While the paper [22] is devoted to

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describing the current scenario and future perspectives, which are essential for strategies of new battery design, recycling routes, and reverse logistics, the second publication [23] reviews the developments of recycling active cathode materials by using various leaching techniques. The different organic acids used to extract metals from spent LIBs have been discussed in terms of their mechanism, efficacies, and other factors (selectivity, cost, etc.) that have to be considered during battery recycling. According to the conclusions of this work, the GHG emissions of Co extraction based on the organic acids stand at 1/8 of the total emission produced using an inorganic acid leaching process.

A further ecological approach devoted to recycling LIBs cathode active materials (especially Co and Li) by using organic acid leaching was investigated by Nayaka et al. [24]. Spent LIBs were generally physically separated; they were subjected to degradable organic acids such as nitrilotriacetic acid (NTA), adipic acid (AA), and ascorbic acid in a time window lasting approximately 6 h. The main benefit of this procedure is that organic compounds are based on different acid concentrations in distilled water; thus, this process produces fewer emissions and hazardous gasses and influences the environment less. Under the optimal process conditions by atomic absorption spectrophotometry (AAS) determination, 75% of Co and 96% of Li ions were leached with NA; 85% of Co and 92% of Li ions were leached with AA via a reductive–complexation mechanism. Moreover, the work analyzes cathode materials' structure and morphology before and after leaching by X-ray diffraction (XRD) and scanning electronic microscopy (SEM).

Eugene et al. [25] introduce a three-stage diafiltration process designed to recycle LIBs for cobalt and lithium higher material recovery. Diafiltration is a continuous green method typically used in operating mode for membrane cascades to achieve recovery of high-purity and high-value products. This paper presents a novel modeling technique and future optimization framework.

Further, Dunn et al. [26] described the environmental burdens, capturing energy consumption, and GHG emissions of the material production, assembly, and recycling of automotive LIBs based on $LiMn_2O_4$ cathode material. In this work, three recycling procedures (hydrometallurgical, intermediate physical, and direct physical recycling) are examined, and the effects of closed-loop recycling on the environmental impacts of battery production are calculated. According to the results of this process-level approach for fully electric vehicles, cradle-to-gate energy corresponds to 75 MJ/kg per battery and GHG emissions of 5.1 kg CO_2e/kg per battery; the direct physical recycling technique can reduce the energy consumption during material production in a closed-loop scenario by up to 48%.

The work by Boyden et al. [27] summarizes the environmental impact of recycling processes from different points of view. Firstly, the introduction devoted to recycling techniques according to their location and efficiency of material recovery is presented. Secondly, the comparative Life Cycle Assessment (LCA) for hydrometallurgy and pyrometallurgy recycling process was established. Due to the environmental impacts in conditions of global warming potential (GWP) 100 (considered over a 100-year period), TETP, and HTP, for these recycling techniques and landfilling of spent LIBs were presented.

The recycling process of spent LIBs is presently constantly evolving and being optimized. The current situation is well summarized in detail in the review work by Bai et al. [28], which, moreover, describes the concept of Battery Identity Global Passport (BIGP). The recyclability of LIBs could be increased by addressing the separation of components using markings, including labels, QR codes, or RFID tags.

Two "ecological" section studies were devoted to the environmental impacts in conditions of China: the work presented by Tang et al. [29] is focused on the social-economic-environmental effects of recycling of spent LIBs from EVs under reward–penalty mechanisms in terms of a Stackelberg game-theoretical model; it considers three different scenarios (no policy intervention, subsidy mechanism, and reward–penalty mechanism). According to the results, even a relatively low minimum recycling rate leads to environmental benefit, consumer surplus, and EV manufacturers' profits. The second work, which Qiao et al. [30] introduced, analyzes the influence of recovered lithium quality on its future availability

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in China. Moreover, it evaluates the potential impact of LIB recycling. The results show, notwithstanding the cell chemistry used, that the recovered lithium would meet roughly 60% in the case of the pessimistic scenario, 53% in the neutral scenario, and 49% for the optimistic scenario of the lithium demand during LIBs production with a recovery rate of 80% by 2050.

3.1.3. Recycling Processes and Recovery Efficiencies

The recycling process covers many important topics. Many publications describe the implemented technique and its effectiveness with a focus on the yield of valuable metals or environmental or economic benefits. Therefore, the initial division of 118 publications into categories: Recovery of Materials, Recycling of Materials, Recycling EV LIBs, and Recycling LIBs were used only in the brief description of the individual publications in Appendix A. Here, the articles were once more divided, and discussed according to the structure shown in Figure 4.

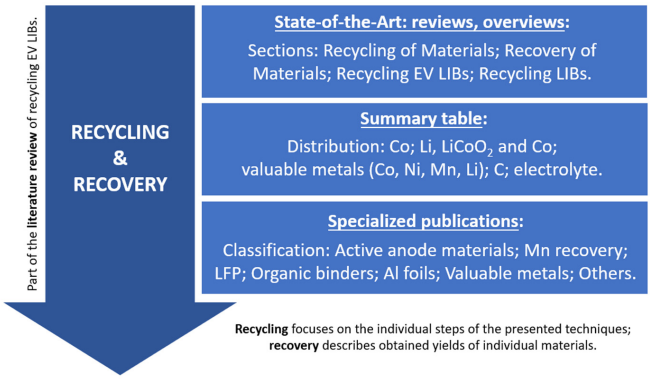

Figure 4. Methodology of Recycling Processes and Recovery Efficiencies caption.

First, the state-of-the-art currently implemented recycling methods and procedures leading to recovery were recorded using published reviews and other overviews. Subsequently, a detailed Table 5 was created. It summarized all selected recycling techniques leading to obtaining Co, Li/$LiCoO_2$/Co, valuable metals (Co, Ni, Mn, and Li), graphite, or electrolyte, describing the procedure, efficiency, or other details. Finally, publications with a different focus were categorized by: Active anode materials, Manganese recovery, LFP batteries, Organic binders, Al foils, Valuable metals, and Others.

Several reviews provide an overall description summarizing the state-of-art devoted to recycling processes and recovery efficiencies of valuable metals. Mansur et al. [31] presented an up-to-date review describing the methods and technologies to recover Co from EOL LIBs. The summary includes thermal, mechanical, and manual preprocessing; the overview of pyrometallurgy and hydrometallurgical methods considers their efficiency levels and future possibilities for improvement. Or et al. [32] devoted their work to covering recycling strategies for valuable metals in mixed-metal LIB cathodes and scrap containing different chemistries. They focused on comparing the environmental footprint and energy consumption between hydro- and pyro-metallurgy. Further, Kim et al. [33] introduced a complex study on recent advances in the anode and cathode materials for the next-generation LIBs including a discussion of the requirements for the high power and energy demands of future energy storage applications, nanostructure synthesis, performance, and reaction mechanisms, and recently used recycling techniques. In another study, Ordoñez et al. [8] presented state-of-the-art recycling technologies for LIBs. Further, Wang et al. [34] discussed the perspective of commonly used recycling processes.

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The work of Garg et al. [35] was focused from a different point of; they described solutions for the screening and regrouping of retired LIBs in terms of secondary application and future recycling. The presented method describes three stages: fast screening technology of voltage and internal resistance, status of health (SOH) detection, and clustering method based on a self-organizing maps (SOM) neural network. Introducing this screening technique would facilitate the identification of the state of the batteries and their subsequent secondary or waste treatment and increase the utility aspect of LIBs. Xie et al. [36] discussed the safety of the recycling process and the consequences of incorrectly chosen disassembly or processing. Emphasis is placed on the current and future work of qualified professional battery recycling enterprises. Further, Yu et al. [37] provided a short overview of the framework of EV LIBs' recycling, considering the access requirement of EV LIBs, transportation of waste batteries, their classification according to the size of a storage system, necessary disassembly requirements, techniques of material recycling, and current EV pollution control.

Piątek et al. [38] critically assessed published research articles and patents on the sustainability of LIBs recycling technologies. The work is devoted to characterizing individual processes, including their potential toxicity or energy consumption, causing CO_2 emissions, and debating the variances in respect of linear and circular economy principles. Moreover, this well-written review presents currently used solutions and future development of the LIBs' recycling field from dismantling over separation to used bioderived materials. A brief clarification for valuable materials recovery from spent LIBs based on pyrometallurgy, hydrometallurgy, and biometallurgy recycling is described by Garole et al. [39]. This review can help select suitable methods for metal recovery and future repurposing. In work by Werner et al. [40], currently used recycling techniques for spent LIBs are described and categorized according to state-of-the-art schemes of waste treatment technology; therefore, the individual units of the process stages are characterized in detail. Huang et al. [41] reviewed contemporary advancements in recycling technologies of spent LIBs, considering the developments in recycling processing, the quality and quantity of obtained products, and the process's effects from the environmental perspective. Azhari et al. [42] discussed the recycling aspects for ASSBs and compared the processes to LIBs' treatment. The work describes in detail solid-state electrolyte chemistries and offers a strategy for ASSB recycling, using hydrometallurgy and direct recycling methods.

Because the previous study focused on Co recovery due to its economic benefit, Liu et al. [43] highlighted the recovery field for Li. They emphasized and evaluated the possibility of industrial realization of each method. Moreover, they discussed these procedures concerning Li's recovery. According to the review: the hydrometallurgy process reclaims Li in the last step; thus, its recovery rate is poor; the pyrometallurgy method lost all the Li in the slag phase (thermal treatment). The optimal method for recovering the most Li is the mild recycling (cleaner production) method, which decreases the temperature of thermal treatment and acid/alkaline step. Li et al. [44] also addressed this issue when they briefly discussed the adsorptive behavior, synthetic methodology, and prospects of sorbents, including spinel LMO, LTO, lithium aluminum layered double hydroxide chloride ($LiCl{\cdot}2Al(OH)_3$). Meng et al. [45] introduced a comprehensive review of Li recovery devoted to industrial practice in perspective to explore technologies for sustainable recovery of Li from minerals, brines, and LIBs.

Other very detailed summaries are provided by Zheng et al. [7], who describe recycling processes for metal extraction such as pyrometallurgy, hydrometallurgy, biometallurgy, and so forth; Zhao et al. [46], who discussed leading technologies and issues in the disposal of spent LIBs from EVs; Gaines et al. [47] brought insight into profitable recycling of LIBs containing a low contribution of Co, while considering new process developments. In other works, Natarajan et al. [48] described current recycling strategies for spent LIB cathodes, Mayyas et al. [49] overviewed challenges in the material supply for automotive LIBs, and Kurz et al. [50] introduced the global warming potential of a new waterjet-based recycling process for cathode materials of LIBs.

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There are also more specialized publications that could be included in other categories of this review, such as a paper by Golroudbary et al. [51] that presented an environmental analysis of the recycling of critical minerals from spent LIBs (more oriented to the overall yield of materials recovery). Further, the work from Tan et al. [52] described the importance of recovering critical materials and improving battery designs from the cell to module level to facilitate recyclability, including the economic and environmental implications. Thyabat et al. [53] showed that applying minerals processing operations decreases the volume of LIBs and NiMH scraps; thus, the leachate purification in the hydrometallurgical process is reduced.

Table 5. Overview of the currently used recycling processes and recovery efficiencies.

Section	Recovered Material	Process	Description	Ref.
Cobalt (Co)	Co	Synthesis of Co_3S_4	Extraction of Co and Ni from the pregnant leach solution (PLS) by a xanthate complex; ammonia solution wash; heat treatment (250 °C, 1 h).	[54]
	Co(II)	Co(II) extraction from chloride using toluene diluted Cyphos IL 102	Co(II) transferred into the organic phase; stripped by 0.05 mol L^{-1} HCl with 99.9% efficiency in a single stage at O/A 1/1.	[55]
	Co, Cu	Electrodeposition	The instantaneous nucleation mechanism occurs at pH 2.7 and progresses at pH 5.4 for Co electrodeposited multilayer on platinum, vitreous carbon, and Al; the same for Cu electrodeposited on Co.	[56]
	Co, Ni	Solvent extraction	Efficient H_2SO_4 + H_2O_2 leaching assisted by diluent heptane and ammonium thiocyanate; reaching factor 372.	[57]
	Co	Carbothermal reduction, magnetic separation	Separation to 53 µm fraction; carbothermal reduction (5–45 min, 500–900 °C); distilled water leaching; magnetic separation of 90% Co.	[58]
	Co	Electrochemical reduction	Molten salt fluidized cathode technique; characterization: voltammograms, chronoamperometry; efficiency 70–80% for the commercial $LiCoO_2$ and upwards of 80% for the spent Li-ion battery.	[59]
	Co, Ni	Leaching	Acid leaching using H_2SO_4, HNO_3, HCl, 1–4 mol L^{-1}, 3–18 h, 25–90 °C, with a solid to liquid ratio fixed at 5% (w/v); the recovery yields of Co and Ni are 100% and 99.99%.	[60]
	Co	Precipitation	The Co(II) hydroxide precipitation; optimal at pH = 9; Co recovery is close to 100% and the filtration flow rate is high.	[61]
	β-$Co(OH)_2$, Co_3O_4	Precipitation, calcination	Chemical (CP) and electrochemical precipitation (EP); Co_3O_4 formation by heat-treating β-$Co(OH)_2$ at 450 °C for 3 h.	[62]

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Table 5. *Cont.*

Section	Recovered Material	Process	Description	Ref.
Cobalt (Co)	$Co(OH)_2$, Co_3O_4	Leaching	The $Co(OH)_2$ is electrodeposited onto conductive glass using −0.85 V, 20 C cm^{-2}, with an efficiency of 66.67%; Co_3O_4 is obtained by heat treatment at 450 °C after 3 h, with an efficiency of 64.29%.	[63]
	Co	Leaching	Acidic dissolution of $LiCoO_2$; next electrodeposition on steel to Co_3O_4.	[64]
	Co	Synthesis	Recovery using 3D sea-urchin-like cobalt nitride composite material (CoN-Gr-2) used as a bi-functional catalyst for water splitting; potentials of 128.9 mV and 280 mV for hydrogen evolution reaction (HER) and oxygen evolution reaction (OER), respectively.	[65]
Lithium (Li)/Lithium Cobalt Oxide ($LiCoO_2$)/ Cobalt (Co)	Li	Separation from solution	The maximum uptake range 20–25 mg Lig^{-1} reached for Amberlite IR 120 resin and molecular sieve 13X.	[66]
	Li_2CO_3, Li_3PO_4	Precipitation	Two-stage precipitation process using Na_2CO_3 and Na_3PO_4, with recovery rates of 74.72% and 92.21%, respectively.	[67]
	$LiCoO_2$	Metal-based leaching	Leaching by Co^{2+} or Mn^{2+}; 95% Li recovery rate and no metal-ions in the leachate.	[68]
	$LiCoO_2$	Eco-friendly leaching	Resynthesis of cathode materials using oxalic acid; 90.13% purity of Li.	[69]
	$LiCoO_2$	Leaching	Using H_2SO_4 and HCl as leaching agents; optimal 2 M HCl, 60–80 °C, for 90 min.	[70]
	Li_2CO_3	Low Li high-salt solution	Li precipitation by P; Li_3PO_4 anolyte dissolution, electrodialysis with cation-exchange membranes used for Li, and P separation with P/Li mass ratio 0.23; Li_2CO_3 precipitation rate reached 88.3%.	[71]
	$LiCoO_2$	Structure restoration	$LiCoO_2$ powder and Li salts sintering to layered structure; Li_2CO_3 addition; calcination at a temperature of 800 °C; coating with nanosized Al_2O_3 particles for performance improvement.	[72]
	Li, Co	Ultrasonic-assisted leaching	Recovery of 96% Co and nearly 100% Li by using 0.5 M citric acid with 0.55 M H_2O_2, a solid-to-liquid ratio of 25 g L^{-1}, a temperature of 60 °C, 5 h, and ultrasonic power of 90 W.	[73]
	Li, Co	Leaching	Leaching using oxalic acid ($H_2C_2O_4$) at 0.46 M at 100 °C; addition of hydrogen peroxide (H_2O_2) resulted in a 33% of activation energy, and 50% of energy consumption reduction.	[74]
	Li, Co	Leaching	Leaching using biodegradable organic methane sulfonic acid (MSA); recovery efficiencies ~100% for Li, Co.	[75]

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Table 5. *Cont.*

Section	Recovered Material	Process	Description	Ref.
Lithium (Li)/Lithium Cobalt Oxide ($LiCoO_2$)/ Cobalt (Co)	Li, Co	Hydrometallurgical-electro dialytic method	Batch extraction of 30% (Co), 69% (Li) using 0.1 M HCl by $LiCoO_2$ dissolution; extraction by compartment electro dialytic cells, and cation-exchange membranes; it yielded a recovery of 62% (Li) and 33% (Co), whereas 80% of Co was electrodeposited at the cathode.	[76]
	Li, Co	Leaching	Cathode leaching using a mixture of citric acid (CA), tartaric acid (TA) and ascorbic acid (AA), to recover the metals; almost complete dissolution of Li, nearly 90% dissolution of Co (80 °C, 6 h).	[77]
	Li, Co	Combination of crushing, ultrasonic washing, acid leaching, and precipitation	Crushing with a 12 mm aperture screen; undersize products ultrasonic washing; filtration through a 2 mm aperture; 4.0 M HCl for 2.0 h, at 80 °C leaching; 97% of Li and 99% of Co recovery.	[78]
	Co, Li_2CO_3, graphite	Thermodynamical	Thermogravimetry analysis, oxygen-free roasting, and wet magnetic separation used to transfer $LiCoO_2$ and graphite powders to Co, Li_2CO_3, and graphite.	[79]
	$LiCoO_2$	Leaching, calcination	Separation of Al foil using dimethyl acetamide (DMAC), next the PVDF and carbon elimination by calcining; well-crystallized single phase $LiCoO_2$ without Co_3O_4 synthesized at 850 °C,12 h.	[80]
	$LiCoO_2$	Suspension analysis	System based on NH_4HCO_3, $(NH_4)_2SO_3$, and NaF, where NH_4^+ represents a complexing agent of NH_3.	[81]
	Li, Co	Ammonia leaching	Leaching rate of 91.16% (Co) and 97.57% (Li), using $NH_3{\cdot}H_2O$ 120 g/L, NH_4HCO_3 75 g/L, n (Na_2SO_3), 80 °C, 240 min.	[82]
Valuable metals (Co, Ni, Mn, Li)	Co, Ni, Mn, Li	Smelting reduction	Li was concentrated and recovered in the flue dust as Li_2CO_3 and LiF. The absence of a slag allows a nearly 100% recovery of Co, Ni, and Mn (alloy) and a nearly 100% recovery of Li (in flue dust).	[83]
	Co, Ni, Mn, Li	Smelting reduction	Smelting reduction in a pilot-scale Electric Arc Furnace in two trials; Co, Ni, Mn and Li's yields are 98.2%, 98.4%, 91.5%, and 68.3%, respectively, in Trial I, and 97.9%, 97.7%, 85.3%, and 60,9%, respectively, in Trial II; carbonated water leaching reaches up the purity of Li_2CO_3 to 95.8%.	[84]
	Co, Ni, Mn, Li	Thermal treatment-ammoniacal leaching	Based on the TG-DSC analysis, cathode material calcined at 300 °C and 550 °C in air atmosphere; Ni, Co, Mn, and Li leached out with efficiencies of 98%, 81%, 92% and 98%, respectively.	[85]

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Table 5. *Cont.*

Section	Recovered Material	Process	Description	Ref.
Valuable metals (Co, Ni, Mn, Li)	Co, Ni, Mn, Li, Cu, Al	Wet crushing, screening, and a ternary leaching system	Selective leaching under conditions: leaching time (0–300 min), temperature (40–90 °C), solid-to-liquid ratio (10–50 g/L), and agitation speed (300–700 rpm); almost completely leaching Ni and Cu, hard recovery of Al, leaching for Li (60.53%) and Co (80.99%).	[86]
	Co, Ni, Mn, Li	Leaching and sol-gel method resynthesis	Leaching using 0.4 mol/L DL-malic acid and 0.1 mol/L ascorbic acid; under conditions (70 °C, 30 min, slurry density: 20 g/L), yields: 99.06% (Li), 97.11% (Ni), 96.46% (Co), and 97.22% (Mn).	[87]
	Co, Ni, Mn, Li	Dissolution−chelation mechanism	Leaching by solution (malonic acid, hydrogen peroxide); procedure efficiency reaches 95% (Li), over 98% (Ni, Co, Mn).	[88]
	Co, Ni, Mn, Li	Leaching	Leaching by sulfuric acid leaching liquor (ammonium oxalate, sodium carbonate solution); precipitation using dimethylglyoxime reagent; solvent extraction using D2EHPA; recovery efficiencies as follows: 98.7% (Ni), 97.1% (Mn), 98.2% (Co), and 81.0% (Li).	[60]
	Co, Ni, Mn, Li	Leaching	Recovery from spent $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$; the efficiencies for Li, Ni, Co, and Mn reached 99.7% under the optimized conditions of 1 M H_2SO_4, 1 vol% H_2O_2, 400 rpm stirring speed, 40 g/L pulp density, and 60 min leaching at 40 °C.	[89]
	Co, Ni, Mn, Li	Leaching	Replacing the oxalates/carbonates of the precipitation process with sulfides; reducing the solubility of Li_2CO_3 in the Li precipitation step with ethanol; reaching the recycling ratio of 94.9% (Li), 94.5% (Co), 94.4% (Ni), and 95.5% (Mn).	[90]
	Co, Ni, Mn, Li	Leaching	Selective leaching system of NH_3-$(NH_4)_2CO_3$-Na_2SO_3; for multistage leaching, high recovery of 98.4% (Li), 99.4% (Co), 97.3% (Ni), and a high-purity (>99%) $MnCO_3$ products.	[91]
	Co, Ni, Mn, Li	Hydrothermal	Using $(NH_4)_2SO_3$ as a reductant in a one-step leaching process; recovery of 100% (Co), 98.3% (Ni), and 90.3% (Li).	[92]
	Li, Fe, P, Al	Physical separation	Discharging of spent LFP batteries in 5 wt% sodium chloride solution for approx. 3 h; extended heat treatment time within the temperature range of 240–300 °C; corona electrostatic separation for metallic particles from the nonmetallic particles.	[93]

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Table 5. *Cont.*

Section	Recovered Material	Process	Description	Ref.
Valuable metals (Co, Ni, Mn, Li)	Co, Ni, Mn, Li	Hydrometallurgical	Recycling $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ materials using 0.2 M phosphoric acid and 0.4 M citric acid with a solid to liquid (S/L) ratio of 20 g/L at 90 °C, 30 min; leaching efficiency of ~100% (Li), 93.38% (Ni), 91.63% (Co), and 92.00% (Mn).	[94]
	Co, Ni, Mn, Li	High-temperature calcination, and coprecipitation	Regeneration of a ternary cathode material ($LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$) using leaching, high-temperature calcination, and coprecipitation procedure; above 97.9% transition-metal elements and 81.2% Li element in the LIBs reused.	[95]
	Co, Ni, Mn, Li	Leaching	Recycling $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ (NMC) using water under strong agitation, and pH-adjusted solutions.	[96]
	Co, Ni, Mn, Li	Extraction and co-precipitation	Extraction by D2EHPA in kerosene—100% (Mn), 99% (Co), and 85% (Ni); Li recovery (purity of 99.2%) from the raffinate as Li_2CO_3 by precipitation; organic load phase stripped with 0.5 M H_2SO_4; cathode material directly regenerated from stripping liquor.	[97]
	Co, Ni, Mn, Li	Leaching	Leaching with 1.0 M H_2SO_4 mixed with 0.62 wt% H_2O_2 at a liquid-to-solid ratio of 25.8 mL g^{-1}, 51 °C, 60 min results in ∼100% recovery of Li, Ni, Co, and Mn; after leaching precipitation into $Ni_{0.15}Mn_{0.15}Co_{0.70}(OH)_2$, and Li_2CO_3.	[98]
	Co, Ni, Mn, Li	Leaching	Using of D, L-malic acid for leaching, and as a chelating agent; synthesis of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ through a sol-gel process (no other chelating reagents).	[99]
	Co, Ni, Mn, Li	Leaching, oxalate co-precipitation, and solid-phase reaction	Preparation of precursor: 50 °C, pH of 1.98, the aging time of 24 h; for calcination 850 °C, 12 h.	[100]
	Co, Ni, Mn, Li	Froth flotation	For multiple stages above 95% of NMC111 in the froth product and 95% of LMO in the tailing product separated.	[101]
	$LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$	Hydrometallurgy	Dismantling, crushing, leaching and impurity removing; $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ prepared from the leaching solution via co-precipitation followed by solid-state synthesis.	[102]
	Co, Ni, Mn, Li	Leaching	Two-step leaching of the $LiNi_xCo_yMn_zO_2$; in the first step, Li and Co selectively leached into oxalic acid at the optimal condition of $C_2H_2O_4$, 0.25 M, pulp density, 10%, H_2O_2 dosage, 0.5%, 80 °C, 90 min.; next H_2SO_4, 3.0 M, pulp density, 6%, H_2O_2 dosage, 2%, 60 ∘C, 120 min. performed; approximately 99% of all remaining metals leached.	[103]

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Table 5. *Cont.*

Section	Recovered Material	Process	Description	Ref.
Valuable metals (Co, Ni, Mn, Li)	Co, Ni, Mn, Li	Leaching	Leaching NMC 811 by using hydrochloric acid (37% w., Sigma Aldrich).	[104]
	Co, Ni, Mn, Li	Leaching, thermal treatment	Solvent method for detaching the current collectors; thermal treatment for removing the polymer binders (PVDF using dissolution with N-methyl pyrrolidone); the polymer solution for carbon separation.	[105]
	Co, Ni, Mn, Li	Leaching	Two-step leaching of the exhausted $LiNi_xCo_yMn_zO_2$ with 99% recovery rate; $C_2H_2O_4$, 0.25 M, pulp density, 10%, H_2O_2 dosage, 0.5%, 80 °C, 90 min for Li, Co; H_2SO_4, 3.0 M, pulp density, 6%, H_2O_2 dosage, 2%, 60 °C, 120 min for other.	[106]
	Co, Ni, Mn, Li Co, Ni, Mn, Li	Leaching	The current collector of Al preforms used as the in-situ reductant of thermite reduction transforming valuable metals of Li, Ni, Co, and Mn were effectively leached into H_2SO_4 solution with efficiencies of 99.78%, 98.62%, 99.29%, and 99.91%, respectively.	[107]
	Li, Fe, P	Sintering	A direct regeneration from spent $LiFePO_4$ batteries using a solid phase sintering; after dismantling, the cathode plate is soaked in DMAC (30 min, 30 °C, and solid-liquid ratio of 1:20 g mL^{-1}); next regeneration at 600–700 °C.	[108]
	Co, Ni, Mn, Li	Calcination, Dissolution	Scraps regeneration suing solvent dissolution and heating at 800 °C.	[109]
	Co, Ni, Mn, Li	Leaching	Method including sol−gel method for resynthesis, and lactic acid (leaching and chelating agent); under 1.5 mol L^{-1}, solid/liquid ratio of 20 g L^{-1}, 70 °C, H_2O_2 content of 0.5 vol%, reaction for 20 min, the results: 97.7% (Li), 98.2% (Ni), 98.9% (Co), and 98.4% (Mn).	[110]
	Co, Ni, Mn, Li	Leaching	The recovery process is based on ammonia, ammonium carbonate, and ammonium sulfite. Co and Cu are completely leached out (~100%), whereas Mn and Al are hardly leached (<10%), and Ni with moderate leaching efficiency (30–50%).	[111]
	Co, Ni, Mn, Li	Hydrometallurgy	Process for recovery $LiNi_xMn_yCo_zO_2$ by applying closed loop recycling; recovery of active cathode material states for over 70% of the battery value.	[112]
	Co, Ni, Mn, Li	Hydrometallurgy	Selectively precipitation using dimethylglyoxime reagent, D2EHPA, ammonium oxalate solution, and saturated sodium carbonate solution. Recovery efficiencies as follows: 98.7% for Ni, 97.1% for Mn, 98.2% for Co, and 81.0% for Li under optimized conditions.	[113]

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Table 5. *Cont.*

Section	Recovered Material	Process	Description	Ref.
Valuable metals (Co, Ni, Mn, Li)	Co, Ni, Mn, Li	Mechanical treatment, Chemical leaching	Mechanical treatment for recovering fractions: ferrous metals, non-ferrous metals, and electronic powders; leaching using Cyanex 272 for Ni and Co, D2EHPA for Mn.	[114]
	Co, Ni, Mn, Li	Leaching	The sulfuric acid was combined with H_2O_2 as a reducing agent; above 99% of valuable metals at 2 M H_2SO_4, 10 vol.% H_2O_2, 75 °C, 300 rpm agitation speed, 250 g/5 L solid/liquid ratio, and after 75 min, was recovered. More than 99% of Li and less than 1% of Co were dissolved at 3 M oxalic acid, at 80 °C, 300 rpm agitation speed, 50 g/L initial solid/liquid ratio, and in 90 min.	[115]
	Co, Ni, Mn, Li	Calcination, solvent extraction, fusion	The 5 h long calcination at 500 °C, solvent extraction aimed at 90 wt% recovery yield for Li salts; the H_2SO_4 and H_2O_2 evaporation resulted in the high purity Co and Mn sulfates; fusion with $KHSO_4$ at 500 °C for 5 h.	[116]
Graphite (C)	C	Leaching and calcination	Sulfuric acid curing-acid leaching; sequential calcination at 1500 °C, where the XRD, Raman spectroscopy, and SAEM analysis were used; final purity of regenerated graphite around 99.6%.	[117]
Electrolyte	PF_6, PO_2F_2, P, F	Transcritical extraction	Combination of extraction and separation; products of hexafluorophosphate (PF_6^-), fluoride (F^-), and difluorophosphate ($PO_2F_2^-$) were detected by ^{19}F and ^{31}P.	[118]

The rest of the publications were devoted to specialized methods describing the possibilities of application of reused outputs, recovery of various materials, or characterizing the selected state. They are described individually in the following sections.

Active Anode Materials

Part of the publications deals with the issue of active anode materials. Yang et al. [119] presented a study of roasting treatment for removing impurities of a spent carbon cathode (SCC) and its application as the anode material used in terms of LIBs. The obtained anode shows a reversible capacity of 365.5 mAh g^{-1} at 0.1 C after 100 cycles with a decay rate per cycle of 0.028%. In a work published by Wei et al. [120], a preposition strategy for silicon/carbon nanofibers/carbon (Si/CNF/C) composite for LIBs, based on micron-sized Si and waste high-density polyethylene (HDPE) as raw materials, was introduced. According to the results, the composite provides an initial coulombic efficiency of 82.2% and a reversible capacity of 937 mAh g^{-1} (1685 mAh g^{-1} based on Si) after 100 cycles at a current density of 100 mA g^{-1}. In the following study, Shen et al. [121] prepared a carbon paper coated with recycled Si and pitch powder (CP-RSP). For the stabilization, 250 °C in air and carbonization at 1000 °C in an N_2 atmosphere were used. The obtained CP-RSP represents the role of the current collector and the active anode material of LIBs. Electrodes with 2.5, 5, and 10 wt.% Si exhibited capacity increases of 94, 129, and 41%, respectively (compared to the silicon-free electrode). Huang et al. [121] investigated the possibility of reusing spent graphite as anode material for LIBs and SIBs. The recovered graphite delivers a capacity of 427.9 mAh g^{-1} after 200 cycles at 0.5 C and displays an outstanding

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rate capability (a capacity of 114.9 mAh g^{-1} is achieved at 3 C). Further, according to Ruan et al. [122], the recovery graphite from spent LIBs was used as a carbon carrier and doped with N and Fe via simple pyrolysis with polyaniline and iron salt. This graphite was used to prepare the ORR electrocatalyst applied in fuel cells. In another work, Bai et al. [123] separated the electrode materials from their current collectors using ethylene glycol; the recovered collectors were intact without corrosion. In another case, Corneal et al. [124] developed used acid baths to separate the active anode materials from the Co foils. Complete separation was reported within 35 s by using 0.5 mol/L of sulfuric acid (H_2SO_4) and a temperature of 40 °C.

Manganese Recovery

In other cases, studies have focused on the recovery of Mn and its compounds. Keller et al. [125] performed solvent extraction of Mn in a lab-scale DN50 pulsed disc and doughnut column. For optimal conditions, 100 g/L D2EHPA (di-(2-ethylhexyl) phosphoric acid) was used as a liquid ion exchanger. In performance tests with 0.01 mol/L $MnSO_4$ solution, a maximum extraction yield of 94% Mn was achieved. Using $LiAlO_2$ seems suitable for recovering Li from LIBs slags [126]. Thus, Wittkowski et al. [127] focused on the characterization of slags of the system Li_2O-CaO-SiO_2-Al_2O_3-MgO-MnO_x, including up to 17 moL% MnO_2 content. According to their research based on plasma optical emission spectrometry (ICP-OES), X-ray diffraction (XRD), electron probe microanalysis (EPMA), etc., the Mn-rich grains are presented as idiomorphic and relatively large (>50 µm) crystals. Wang et al. [128] examined the direct recycling process for a cathode scrap; their procedure is based on sulfate radical-based advanced oxidation processes (SR-AOPs) and provides a complex synthesis. The results show that the cathode scrap includes high efficiency for peroxymonosulfate (PMS) activation, whereas the ortho-phenyl phenol (OPP) degradation still reached 94.8% after ten cycles. In conformity with heterogeneous catalyst thermal regeneration strategies, Poyraz. et al. [129] provided a sustainable manganese-based material, where binder-free self-supporting (BFSS) electrodes use a fibrous, high aspect ratio MnO_2 active material. After 200 discharge–charge cycles, the reuse-based BFSS electrodes showed crystallinity and oxidation state of the manganese centers on a similar level as new BFSS cathodes.

Lithium Iron Phosphate (LFP) Batteries

A wide range of current research on lithium-ion battery technology focuses only on the LFP type. The same was the case with Shin et al. [130], who presented a green process to recycle $LiFePO_4$/C electrode materials by using a precursor of a crystalline $FePO_4 \cdot 2H_2O$ phase (metastrengite I). According to their calcination heat treatment, cathode materials are synthesized at 700 °C and deliver a maximum discharge capacity of 168.51 mAh g^{-1} at 0.1 C with a capacity retention of 99.36% after the 25th cycle at 1 C. The rest of their study compares their results to the commercially available $LiFePO_4$ powders. An approach for reusing the spent LFP electrodes was introduced by Gangaja et al. [131]. Regenerated $LiFePO_4$ as LIB half-cells reveal a capacity of 145 mAh/g at 1 C and 107 mAh/g at 10 C and 96% capacity retention at 5 C for 300 cycles. Other experiments performed by Li et al. [132] were devoted to direct recycling; through this procedure, high purity of cathode material, including $LiFePO_4$ and acetylene black, anode material based on graphite and acetylene black, and other process by-products such as shell, Al foil, Cu foil, and electrolyte solvents were obtained. The regenerated cathode material mixture regenerated at 650 °C, meeting the reuse requirement for middle-end LIBs. In the procedure presented by Liang et al. [133], spent LFP cathode material was regenerated through Li/Fe/P elements compensation, and its structure was reshaped via the heat treatment method in the temperature range of 450–650 °C. Given their results, the material is restored to the initial conditions; its surface and particle sizes are smooth and sufficiently small.

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Organic Binders

Organic binders play a crucial role in LIBs' composition; they are responsible for the active material particles' strong connection between the electrodes and the metal contacts. Regardless, their use causes significant difficulty in extracting pure electrode materials. High-temperature treatment such as pyrolysis or incineration is being executed for their removal and separation; this approach leads to fluorinated exhaust gas emissions. Fu et al. [134] devoted themselves to extraction based on supercritical carbon dioxide (SC CO_2) combined with a cosolvent dimethyl sulfoxide in the case of liberation of the cathode materials from Al foil. The results show that 98.5 wt% polyvinylidene fluoride (PVDF) dissolves in the SC CO_2 dimethyl sulfoxide system under the optimum conditions: 70 °C, 80 bar pressure after 13 min. In another approach introduced by Hanisch et al. [135], the PVDF binder thermal decomposition was performed to weaken the adhesion between coating and foil. Moreover, they provided a laboratory-scale ANVIIL separation process (Adhesion Neutralization via Incineration and Impact Liberation). According to their results, 97.1% w/w of the electrode materials were regained with Al impurities 0.1% w/w.

Aluminum (Al) Foils

Other studies focused on the processing of Al foil. Chu et al. [136] separated positive active materials from Al foil, while the foil can be kept intact. This procedure targets prior high Li recovery. The examination of the ultrasonic-assisted acid scrubbing method for coating materials and Al foils separation was published by Chen et al. [137]. About 99%, 100%, and 46% coating materials can be detached in sulfuric acidic, oxalic acidic, and pure water medium, respectively, whereas the Al foils had a corresponding purity of 98%, 99%, and 15%.

Valuable Metals

Recovered valuable metals (Co, Li, Mn, and Ni) are reused in producing the active materials for the new LIBs. Yang et al. [138] dealt with preparing the next-generation high-performance phosphate cathode materials and examined a high voltage/energy and long-life $LiFe_{0.6}Mn_{0.4}PO_4$/C (LFMP/C) composite. It offers a capacity of 141.3 mAh/g at 0.1 C and 104.9 mAh/g at a high rate of 10 C, and shows a long cycle life, e.g., 86.4% capacity over 400 cycles at 1 C. Further, Sloop et al. [139] introduced an approach to producing low-cost, recycled, battery-grade electrode material. This soft-chemical treatment leads to NCM 523 and NCM 622 LIBs with a performance equivalent to the originally manufactured ones. Zheng et al. [140] exhibited four representative recycling streams to generate consistent quality cathode material of NMC111. The precursors and the cathodes show similar morphology, particle size, and tap density; the capability of recovered NMC111s was better than for a commercial NMC111.

Others (Not Specified)

The following publications were devoted to specialized, unclassified research: Liu at el. [141] proposed the reuse of Si and lignin waste from photovoltaics (PV) and the traditional paper industry to fabricate high-capacity silicon/carbon (Si/C) anode materials for LIBs using electrostatic attracting force and thermal process. The obtained Si/C composite displayed an initial charge capacity of 1016.8 mAh/g, a high-capacity retention of 74.5% at 0.2 A/g after 100 cycles. Hou et al. [142] recycled waste Cu scraps in the form of CuCl powders via the facile hydrothermal route considering its high economic value added (EVA) characteristic. After a series of characterization procedures, including scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDX), and X-ray diffraction (XRD), the reversible discharge capacity of CuCl powders as the active anode material was determined. It was about 171.8 mAh/g at 2.0 C even after 50 cycles. Zhang et al. [143] studied the adsorption of spent LFP and LMO cathodes as adsorbents toward heavy metals in water in terms of adsorption time, initial adsorbate concentrations, and co-existing ions on adsorption kinetics. According to the results, LFP adsorption capacities were

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44.28, 39.54, 25.63, and 27.34 mg g^{-1} for Cu^{2+}, Pb^{2+}, Cd^{2+}, and Zn^{2+}, respectively; in the case of LMO, similar adsorption capacities were achieved (32.51, 31.83, 26.24, and 25.25 mg/g, respectively).

In a work by Siebenhofer et al. [144], the liquid membrane permeation with supported flat sheet membranes was examined. The stability and accuracy of supported liquid membranes (SLM) for LIBs materials were evaluated; it was performed in accordance with the HDEHPA test system. The optimum concentration for the transport of Co through the SLM with LIX 84 (hydroxy-5-nonylacetophenonxime) was found to be 10 wt.%. The other work of Zhang et al. [145] was devoted to the pyrolysis kinetics of cathode material using various methods, including Flynn–Wall–Ozawa (FWO), Friedman, Kissinger–Akahira–Sunose, Starink, Tang, and Boswell. The Coats–Redfern inspected the thermal degradation mechanism. In compliance with the thermogravimetric analysis, three stages of mass losses of cathode material decomposition (1.51%, 0.787%, and 0.449%) were established. Kim et al. [146] investigated using collected silicon oxides (SiO_x) condensed particles from Si vapors from the ingot-growing furnace as another method for green production of active anode material for LIBs. Different characterization analyses, such as FE-SEM, TEM, EDS, XRD, and XPS, were completed. A cycle with 40.6% efficiency of the produced material was determined under charge and discharge tests in 100 cycles, 0.2 C for the first three cycles and 1.0 C for the remaining 97. Further, Chen et al. [147] converted the recycled LCO from spent LIBs into an efficient electrocatalyst for oxygen evolution reaction (OER). After 500 cycles, a current density for the electrocatalyst of 9.68 mA cm^{-2} at 1.65 V was determined. Beheshti et al. [148] introduced secondary Al production as an acceptable process for recycling spent LIBs. The study focused on recovering Al, Co, and Li from mixed waste streams and Al scrap.

In another work, Hoshino et al. [149] developed a procedure for Li recovery from seawater by electrodialysis using a Li separation membrane with the ionic liquid PP13-TFSI. This method suits seawater desalination and Li dissolution regarding recycling spent LIBs. Bae et al. [150] devoted their research to the waste-to-lithium system based on an electrochemical reaction with water and Li precursors, such as LiOH and Li_2CO_3, executed at room temperature. The obtained Li metals have a high purity (over 99%), and the produced Li_2CO_3 is phase-pure without any remarkable secondary phase. Sobianowska-Turek et al. [151] discussed the situation in the market of portable LIBs in the European Union (EU), focusing on Polish waste treatment management systems and disposal solutions. The work critically described current problems and deficiencies in the approaches to the end-of-life procedures for waste batteries and accumulators in the conditions of Poland and introduced possible future solutions. Peng et al. [152] studied metals' purification, recovery, and reuse from spent LIBs with high efficiency and low costs. During their experiments, they removed target impurities from a solution individually: Fe and Al were removed by changing the pH value; Co was purified using selective electrodeposition technology and solvent extraction. Next, they co-precipitated obtained products and synthesized them for the final comparison of the LNCM-R and LNCM-N. Renault et al. [153] introduced the environmentally friendly process of recycling Li from organic electrode materials for secondary used LIBs. Over 99% of the recovered material's capacity can be reached by this process when comparing the second applied battery with the original one. The results were achieved by a dimethyl carbonate/lithium bis(trifluoromethanesulfonyl)imide electrolyte. Asari et al. [154] provided a chemical analysis questionnaire survey and flow analysis results regarding Co recovery under LIBs recycling conditions in Japan. Jo et al. [155] initiated an efficient direct physical and combined metallurgical recycling procedure to lower LIBs' environmental burden. The electrochemical performance of the active electrode material based on the recovered materials is similar to commercially available ones. Rouhi et al. [156] performed a systematic work devoted to the behavior of LIBs during discharge in salt solutions. The result shows that the voltage recovery effect is not stable and adequate and can cause risks during further processing. Moreover, the methodology to lower open

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circuit voltage in an aqueous salt solution to 2.0 V, which is acceptable for mechanical processing, is provided.

3.1.4. Life Cycle Analysis (LCA) Study

Life Cycle Assessment, also known as Life Cycle Analysis (LCA) study, represents a methodology designed for assessing environmental impacts during all the stages of the life cycle of commercial products, processes, or services. It describes environmental aspects and potential consequences that are assessed during the raw material extraction and processing (cradle), through manufacturing, distribution, and use, to the End-of-Life (EOL) activities such as recycling and final disposal of the material compositing (grave). The LCA stages diagram is illustrated in Figure 5. The main aim of the LCA study is to document and provide the lifelong overall environmental profile of the selected product [157,158].

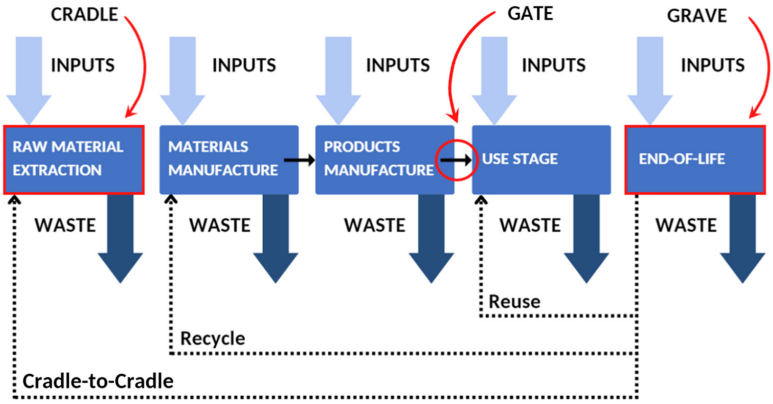

Figure 5. LCA stages diagram including Cradle-to-Grave loop.

Many attempts were conducted to standardize LCAs to deliver an objective and comparable result in study cases. Nowadays, widely recognized procedures for assessing LCA are included in the 14,000 series of environmental management standards of the International Organization for Standardization (ISO) in:

- ISO 14,040—that provides the 'principles and framework' of the Standard, and in simple terms, is written for a managerial audience,
- ISO 14,044—that offers concepts of the 'requirements and guidelines' typically used by practitioners [159].

On the contrary, limiting and introducing one unique LCA method would reduce the quality of the obtained results, and the complete assessment would be affected and not comprehensive. Therefore, the standards describe two main types of approaches:

- Attributional LCA—attempts to answer 'how and which impacts are flowing within the chosen temporal window?',
- Consequential LCA—tries to answer 'how will they flow beyond the immediate system change in response to our decisions?' [160].

As it is clear from the definition and characteristics of LCA, the studies are not performed at the same level; therefore, they cannot be mutually compared. The executed methodology, the boundaries case set, and the primary work intent are considered for each LCA individually. A total of five publications dealing with life cycle issues of LIBs from EVs were evaluated using the performed literature search (three Articles and two Proceedings Papers.

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A comprehensive and complex work devoted to the life cycle assessment of different types of LIBs during their recycling processes was presented by Mohr et al. [161]. They focused on four different cell chemistries—NCA, NMC, LFP, and the emerging sodium-ion battery (SIB) technology. In their work, three different recycling techniques of spent LIBs were set in contrast:

- Two basic kinds of recycling treatments, the pyrometallurgical and hydrometallurgical one, based on secondary inventory data from the current state-of-the-art LCA models,
- One advanced hydrometallurgical technique modeled on the first-hand data obtained from industry.

Considered process flows are shown in Figure 6 and are described in more detail in the complete form of this work [161]. Details of individual technologies of the mentioned recycling processes are summarized in the first part of this literature review devoted to recycling technologies of spent LIBs from EVs [5].

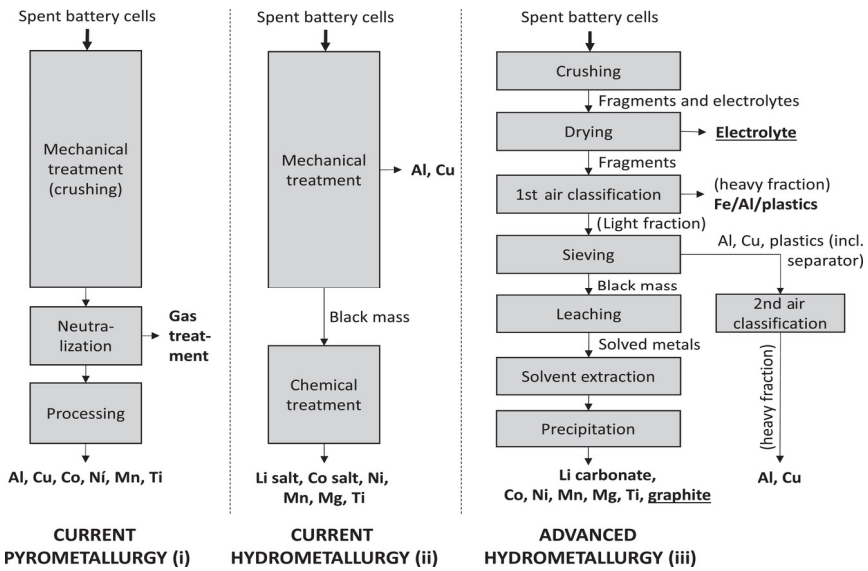

Figure 6. Process flows, including all considered product outputs. Reprinted from Journal of Industrial Ecology, Vol. 24, Mohr et al. [161], Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes, no. 6, pp. 1310–1322, Copyright (2022), distributed under Creative Commons Attribution License.

Obtained results are presented in three branches: production, recycling, and net impact. Because, by interpreting the results, associated uncertainties need to be considered, detailed sensitivity analysis mapping future progression is included [161].

According to that, NMC production causes the lowest GWP per 1 kWh capacity (75.50 kg CO_2-Eq), followed by NCA, LFP LIBs, and SIB. Battery cell manufacturing represents the most significant part of the production GWPs for each chemistry type. The cathode material is a serious producer of the total GWP as abiotic resource depletion potential (ADP) for LIBs (especially NMC and NCA). The highest recycling benefits are obtained for processing components that have a high impact during primary production, such as copper, nickel, cobalt, and, considering the GWP aspects, aluminum. This, and considering ADP aspects, is shown in Figure 7. The advanced hydrometallurgical method presented

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in this work has the highest benefit in all cases due to the additionally recovered graphite and electrolyte [161].

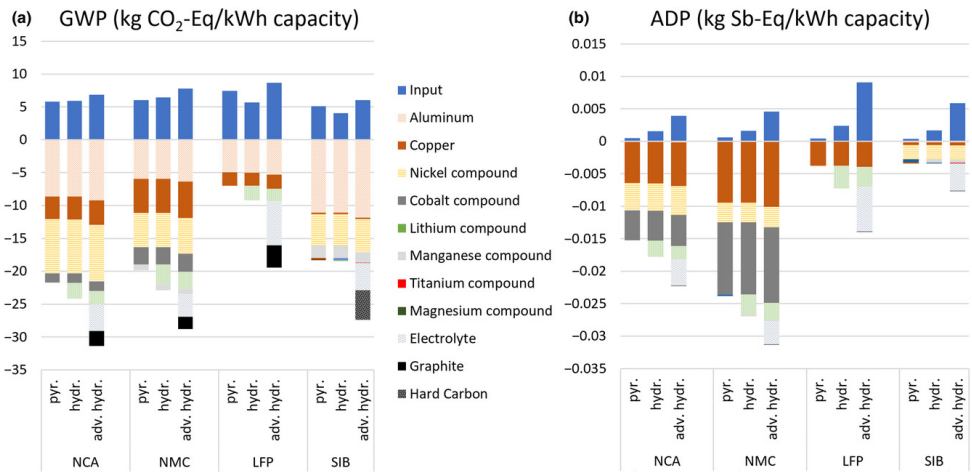

Figure 7. Environmental benefits of battery cell recycling, broken down to the contribution of the different fractions recovered by the recycling processes: (**a**) global warming potential (GWP), (**b**) abiotic resource depletion potential (ADP). Negative values indicate net benefits (reduction of impacts due to recovered materials), positive values are environmental impacts (due to process inputs and emissions, e.g., energy and chemicals, off-gasses). Reprinted from Journal of Industrial Ecology, Vol. 24, Mohr et al. [161], Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes, no. 6, pp. 1310–1322, Copyright (2022), distributed under Creative Commons Attribution License.

Following this assessment, the highest recycling benefits and lowest net impacts for NCM and NCA LIBs were indicated. Benefits for LFP LIBs and SIB reach comparable low levels considering the similar impact criteria. Further, the GWP and ADP production is significantly affected by the procedure of the used recycling method. Moreover, this work includes a short review of existing LCAs on LIBs recycling used as a state-of-the-art base for model parametrization [161].

Unterreiner et al. [162] provided standard-based LCA devoted to the environmental impacts of recycling and material reuse of three types of batteries: lead-acid, LIBs, and vanadium redox flow. In this study, the reusing of materials is considered in a closed-loop production (cradle-to-cradle), where the EOL disposal step for products is replaced by recycling. This method is used to minimize the impacts of the products by employing principles of sustainable production, operation, and practices; it means that the materials that can be reused are counted as a benefit during the LCA study implementation [162].

The presented evaluation is based on Life Cycle Inventory (LCI) Analysis, which includes material production, energy consumption, energy losses, and transport requirements. For the interpretation, the Life Cycle Impact Assessment (LCIA), based on the categories from the ReCiPe2008 method, was used. Study categories were converted to ecological points, including waste management such as recycling, incineration, or landfilling in the recycling phase [162].

The results show that the LIBs have the lowest ecological impact compared to the other examined technologies. Nevertheless, this impact could still decrease by more than 20%. Moreover, around 62% of LIB materials are currently reusable. However, Li is not

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counted and reused due to economic reasons. According to the study, the recycling and reuse of Li would have a small ecological impact [162].

In the study by Gaines et al. [163], the life cycle energy performance analysis of LIBs from plug-in hybrid electric vehicle PHEV-20 was discussed. Due to the results, the environmental impacts of the cradle-to-gate cycle (it means the part of the product life cycle from material extraction to the factory gate but before the transport to the final consumer) is small, corresponding to a few percent. Further, recycling battery materials can potentially reduce the production energy by about 50%. Results are presented in the form of energy data comparable to GHG or can be used further for GHG calculation [163].

Dunn et al. inducted a series of publications [26,164,165] devoted to the environmental perspective of automotive LIBs. These issues are entirely examined in the detailed LCA [164], and potential hot spots within the life cycle for five cathode materials and a lithium anode are identified. Notwithstanding, the NMC cathode material production is over three times more energy intensive than LFP, and its energy density is, on average, 1.5 times higher. The results specify that less cathode material is needed in the battery with the more energy-dense cathode; thus, the overall battery mass is lower. It yields a lower cradle-to-gate GHG and energy intensity. Moreover, this work presents a GHG emission comparison of electric and well-to-wheels conventional vehicles (CV) [164].

The LCA of a new promising type of LIBs for EVs based on lithium cobalt phosphate (LCP) chemistry was executed by Raugei et al. [166]; the study focused on CED and GHG. As with other chemistry types, the cathode is the main contributor to high CED (higher than 60%) and GHG (at least ~70%; this percentage represents the cradle-to-gate scenario). According to the results, the inclusion of the EOL treatment steps leads to a mild marginal reduction of the total CED (2%) and a larger relative reduction of GHG emission (8%) [166].

3.2. Economic Perspective

Few publications deal with the economic questions of LIBs technology; four Reviews, four Proceedings Papers, and six Articles were described as part of this review. A comprehensive view of LIBs within their circular economy (CE) is often evaluated, or techniques' financial costs and returns are assessed and compared. For more detailed analyses leading to the determination of the exact financial burden or benefits of recycling LIBs, e.g., in a selected location or for a designated technology, it will be necessary to carry out many other research steps.

Reusing and recycling of spent LIBs was reviewed in the work by Pagliaro et al. [167], which brings insight into the concept of circular economy (CE). The work simply describes the mechanisms of the recycling process and provides an overview devoted to renewable electricity storage on the world-wide scale. In more detail, the CE is discussed by Mossali et al. [168]; they provided a literature review of current-state opportunities of recycling treatments and, moreover, an overview of valid patents related to industrial recycling processes. Velázquez-Martínez et al. [10] offered an analysis of recycling technologies from a CE perspective focused to the industrial field. The work describes the industrial recycling mechanisms of nine companies and laboratory processing suggested by Aalto University in detail. Moreover, the work provides graphic representations of all processes and discusses the efficiency and quality of the recovery outputs. The companies and their forms of output materials based on Co, Li, and other metals are listed in Table 6. Martins et al. [22] discussed the environmental and economic issues of recycling and future approaches considering their sustainability. The work is devoted to a detailed overview of CE in EVs. Moreover, the correlation between the number of used vehicles and GDP is described. Dalini et al. [169] reviewed the environmental and economic aspects of recycling by a hydrometallurgical technique, including the pretreatment, leaching, precipitation, extraction, and electrochemical methods leading to the regeneration and recovery of valuable metals. Further, Zhao et al. [170] reviewed the state-of-the-art of recycling processes of spent LIBs based on the $LiCoO_2$ system; they discussed the possible problems and prospects according to the recently published works.

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Table 6. Recovery materials of nine recycling companies with their specified form of output. Reprinted and edited from Batteries, Vol. 5, Velázquez-Martínez et al. [10], A critical review of lithium-ion battery recycling processes from a circular economy perspective, no 4., Copyright (2022), distributed under Creative Commons Attribution License.

Process Company	Forms of Output Materials				
	Co	Li	Fe	Al, Cu Foils	Loses
Umicore Valéas™	$CoCl_2$	x	Recovered in alloy	Recovered in alloy	Al, polymer
Sumitomo-Sony	CoO	x	Recovered in alloy	Recovered in alloy	Al, polymer
Retriev Technologies	MeO + cake	Li_2CO_3	Recovered in shaking table	Not specified	Not specified
Recupyl Valibat	LCO/$Co(OH)_2$/Co	Li_2CO_3/ Li_3 PO_4	Recovered with magnetic separator	Al, Cu recovered with density separation	Not specified
Akkuser	Co + graphite	x	Recovered with magnetic separator	Al, Cu recovered from fin powder	Al fraction
Accurec	Co alloy	Li_2CO_3	Recovered via air filtration	Al, Cu recovered via air separator, Al via leaching	Polymer
Battery Resources	$NMC(OH)_2$	Li_2CO_3	Recovered with magnetic separator	Cu recovered via dense media separation and precipitation	Not specified
LithoRec	MeO	Li_2CO_3/ LiOH	Recovered via air filtration	Al, Cu recovered via sieves and zig-zag sifter	Not specified
OnTo	Cathode powder (refurbished)	Li_2CO_3/ cathode	Recovered, not specified	Recovered, not specified	Not specified

Other publications provided a more specific economic point of view. Natkunarajah et al. [171] presented former scenario analyses devoted to the return rates of LIBs and return rates based on the market shares of EVs. They analyzed return rates considering the lifespan of different battery systems in their first life application (EV) and a possible second life application (battery energy storage). The prediction data for batteries of BEV, PHEV, and HEV are based on the current EVs on the German market.

Steward et al. [172] presented different directions for future developments based on the results of techno-economic analyses made for spent LIBs. Their overview discusses savings from recycling cathode materials from the EOL LIBs relative to the production use of virgin materials. Ma et al. [173] estimated the economic profits of recycling spent $LiFePO_4$ (LFP) and $Li(NiCoMn)O_2$ (NCM 523) batteries in the conditions of China. In their work, they calculated recycling revenues (R), determined recycling costs (C), and established profits of the recycling (B) for each battery type. The considered costs and profits have been converted from CNY to USD, according to the current exchange rate of July 2022, and are shown in Table 7. Garg et al. [174] presented an artificial intelligence (AI) approach to thermo-mechanical-electrochemical-based evaluation for the residual energy of the LIBs embedded in battery packs used in EVs. The study's findings are built on a robust model using genetic programming; it can optimize the recycling strategy for spent LIBs and leads to the reduction of the cost of the whole process.

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Table 7. The recycling profits of the spent power batteries. Reprinted and edited from IOP Conference Series: Earth and Environmental Science (EES), Vol. 159, Ma et al. [173], The Recycling of Spent Power Battery: Economic Benefits and Policy Suggestions, no. 1, Copyright (2022), distributed under Creative Commons Attribution License.

Battery Type	Profits (B) (USD/Ton)	Revenues (R) (USD/Ton)	Costs (C) (USD/Ton)
LFP	505	1.57	1.07
NMC 523	2613	5.47	2.85

Lander et al. [4] introduced a techno-economic model for comparing recycling locations and techniques; they presented it as a key tool for recycling cost optimization in an international battery recycling economy. According to this study, recycling of LIBs can be economically viable, with cost/profit ranging from (−21.43–+21.91) USD·kWh^{-1}. However, the final cost/profit sum depends on the form and distance of transport, design of the battery pack, and the recycling method. The study provides six different scenarios according to their location (China, South Korea, US, Belgium, and the UK) performed for five types of LIBs (NCA, NMC 622, NMC 811, LFP, and LMO). The net recycling profit in USD·kWh^{-1} for the recycling of various commercial battery packs is shown in Figure 8.

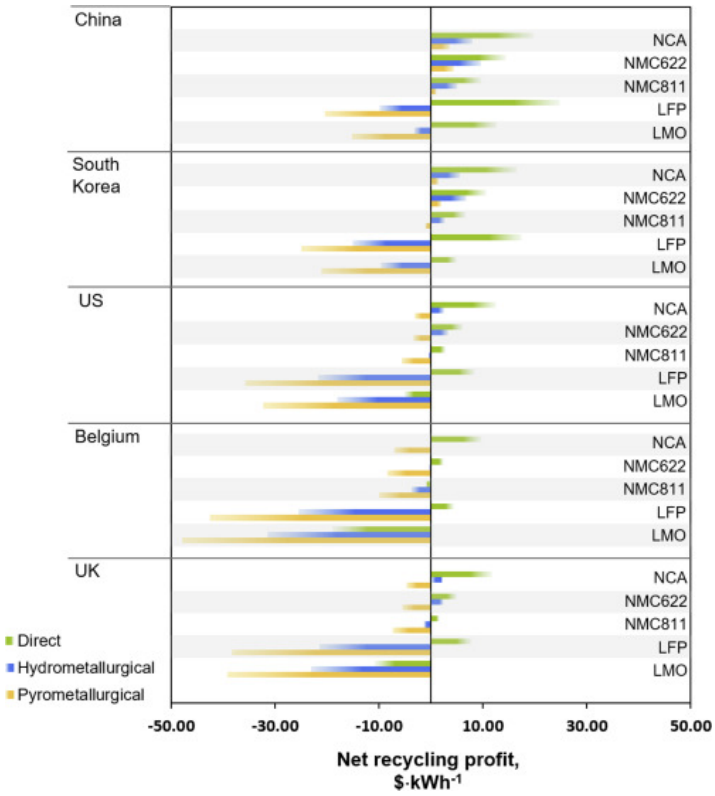

Figure 8. Net recycling profits for commercial EV battery packs. Reprinted from iScience, Vol. 24, Lander et al. [4], Financial viability of electric vehicle lithium-ion battery recycling, no. 7, Copyright (2022), distributed under Creative Commons Attribution License.

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Wang et al. [175] established a model for the profitability analysis of recycling three different types of LIBs (LCO, LFP, LMO) based on the commodity market prices for recovered materials. The profitability is highly dependent on the mix of cathode material chemistries in the waste and the resulting variableness in material mass. Considering the results, the potential values of the waste streams range from USD 860/ton for $LiMn_2O_4$ cathode batteries to USD 8900/ton for $LiCoO_2$. In another work, Li et al. [176] developed an economical green recycling method for recycling spent $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ cathodes; more than 98% of Li, Co, Ni, and Mn was leached out by acetic and maleic acid. The economic analysis was performed considering the costs of leachates (organic acids and reducing agents) and the energy consumption of processing. According to that, the cheapest recycling would be achieved using H_3PO_4 as a leaching acid, costing USD 16.53/kg of cathode material. The cost of used acids is USD 49.82/kg for acetic and USD 94.97/kg for maleic acid, considering the mass of the cathode material. Wang et al. [177] developed an environmental economic model considering CO_2 emission to simulate recycling spent LIBs. It provides three handling strategies: battery recycling, remanufacturing, and disposal. Moreover, a real case study from a Chinese EV manufacturer is introduced. According to the study results, a 5.7% decrease in the total cost and a 21.8% emission reduction can be achieved.

4. Discussion and Conclusions

Lithium-ion batteries are a key technology—from small portable consumer devices, through electromobility, to large-capacity energy storage. The ever-increasing number of EVs on the market, supported by still-stricter requirements for reducing decarbonization and CO_2 emission reduction, brings several requests, including lowering the prices of new EVs or searching for strategies in waste management solutions and processing of spent batteries.

Battery recycling appears to be the most suitable solution. Reusing rare metals (Ni, Co, Mn, or Li) or packaging materials (Al, Cu) limits the extraction of new raw ones and lower energy consumption and emissions. Moreover, implementing the proper process leads to a price reduction of a new battery and the whole vehicle.

This work represents the second part of the literature review based on peer-reviewed publications devoted to "Recycling of Lithium-ion Batteries from Electric Vehicles" and summarized 162 papers focusing on the environmental and economic (E&E) perspective. The reviewed works were classified into five sections: Recycling Processes, Battery Composition, Environmental Impact, Economic Evaluation, and Recycling and Rest, where this part deals in detail with the last four mentioned. The outline of discussed branches is illustrated in Figure 9.

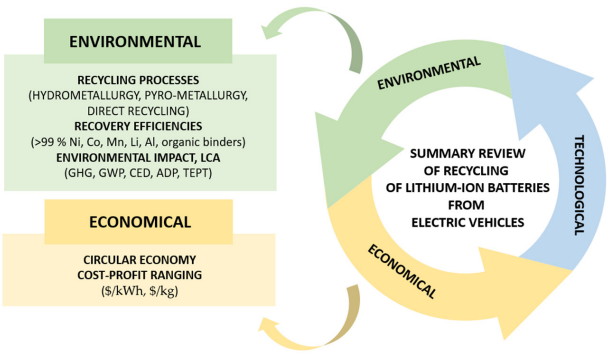

Figure 9. The summarizing figure of the second part of the literature review on recycling lithium-ion batteries from electric vehicles focused on environmental and economic impacts.

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The following can be concluded based on the review:

- Only a few publications are devoted to the recycling of the electrolyte issue. Although the organic electrolyte is the least profitable item of the whole battery (compared to the recovery of high-in-cost metals), it is required to complete studies characterizing its safe waste treatment, toxicity analysis, and environmental impacts, including wastewater treatment procedures or proper disposal.
- Several studies have been conducted addressing the effect of reclaimed materials on a new production of raw materials, either on a small scale or based on laboratory techniques. Nevertheless, it would be beneficial to study this issue further, especially on the data of implemented lines, which could represent a real scenario and outline future possibilities.
- According to Table 5, considering the available recycling processes, more than 99% of all valuable metals (Ni, Co, Mn, and Li), Al coverings, or organic binders can be recovered. Currently, these processes are being optimized to reduce necessary costs or environmental impacts.
- Several comprehensive LCA studies have been conducted, characterizing the issue of LIBs from EVs during their active and waste life. Although most works deal with GHG topics, there are extensions evaluating the effects of TETP (terrestrial ecotoxicity potential) or CED (cumulative energy demand). Including other environmental indicators would be beneficial, such as global warming potential (GWP) or abiotic resource depletion potential (ADP).
- So far, few publications have addressed the economic aspect of recycling LIBs from EVs. Although some complex works can be found, it is necessary to devote further research in this direction to achieve a high-quality evaluation of financial impacts with full use of EVs LIBs.

The first part of this review collection dealt with the technology and commonly used principles of recycling processes on the laboratory (small) and industrial (large) scale. This part brings additional E&E insight into publications considering recycling of spent LIBs from EVs published until 2021. Thus, these works provide a comprehensive techno-environmental economic overview of recycling EVs LIBs, which can be further used in academic and industrial spheres as a starting point for knowledge of this topic.

Author Contributions: A.P.: conceptualization, methodology, resources, data curation, writing—original draft preparation, visualization, project administration; V.K.: writing—review and editing, supervision, project administration; D.-I.S.: writing—review and editing, project administration. All authors have read and agreed to the published version of the manuscript.

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Appendix A

Table A1. Assessed literature in review for "Cathode", "Anode", and "Electrolyte" category.

Battery Cell Composition			
Reference	**Type**	**Publication Year**	**Summary Content**
[11]	Review	2020	The recycling process of anode materials and electrolyte; recently used techniques; recovery materials application.
[12]	Proceedings Paper	2012	Summary of recycling techniques used for LIBs.
[13]	Proceedings Paper	2005	Discussion of recycling techniques used for LIBs.
[15]	Proceedings Paper	2018	Direct recycling of bare and coated NMC 622 cells.
[14]	Article	2021	Strategy for recycling LFP batteries; cathode composition based on recycled materials, design of new-type dual-ion battery.
[16]	Article	2021	Physical separation/recycling process, implemented via thermal and mechanical treatments on LFP cathode materials and current collectors (including Al fragments).

Table A2. Assessed literature in review for "Envi/Ecological Impact" category.

Environmental and Ecological Point of View			
Reference	**Type**	**Publication Year**	**Summary Content**
[23]	Review	2020	Current-state developments of recycling active cathode materials by using various leaching techniques (organic acids).
[25]	Proceedings Paper	2019	Three-stage diafiltration process for recycling LIBs (Co, Li).
[27]	Proceedings Paper	2016	Summarization of recent recycling techniques; LCA; environmental impacts (GWP, TETP, HTP).
[18]	Article	2009	Critical analysis of natural resource savings.
[19]	Article	2017	Environmental and economic impacts of reusing recovery material during new LIB production.
[20]	Article	2014	Comprehensive analysis of energy and environmental impacts devoted to material production and battery assembly; motivation for recycling LIBs if considering high energy consumption of assembly; comparison EVs vs. ICVs.
[21]	Article	2021	The dis/advantages of currently used recycling techniques for LIBs from EVs; environmental issues of EVs' LIBs' production, use, and EOL procedures.
[22]	Article	2021	Current scenario and future perspectives for LIBs recycling; correlation between the vehicles in use and GDP.
[24]	Article	2018	Ecological recycling cathode materials (especially Co, Li); organic acid leaching (NA, AA, ascorbic acid).
[26]	Article	2012	Cradle-to-Gate energy consumption and GHG emissions of LIBs from EVs.
[28]	Article	2020	Summary description of current recycling techniques;concept of Battery Identity Global Passport (BIGP).
[29]	Article	2019	The social-economic-environmental impacts of spent LIBs from EVs; three scenarios for Stackelberg game theoretical model.
[30]	Article	2020	Determination of the future lithium availability in China.

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Table A3. Assessed literature in review for "LCA" category.

Life Cycle Assessment (LCA)			
Reference	**Type**	**Publication Year**	**Summary Content**
[162]	Proceedings Paper	2016	LCA (cradle-to-cradle) for lead acid, LIBs, and vanadium redox flow based on ReCiPe2008 method.
[164]	Proceedings Paper	2016	LCA of cathode materials (NMC, LFP, three types: LMR-NMC), and a lithium anode; GHG comparison EVs and CVs.
[161]	Article	2020	LCA for NCA, NMC, LFP, and SIB; comparison of basic pyrometallurgy, hydrometallurgy, and advanced hydrometallurgy; evaluation using GWP, ADP.
[163]	Article	2011	Life cycle energy performance analysis—PHEV-20.
[166]	Article	2019	LCA of LCP battery, focused on CED and GHG.

Table A4. Assessed literature in review for "Recovery of Materials" category.

Recovery of Materials			
Reference	**Type**	**Publication Year**	**Summary Content**
[43]	Review	2019	Summary of Li recovery procedures and their evaluation.
[44]	Review	2018	A brief review of Li recovery from aqueous resources.
[45]	Review	2019	A comprehensive review of Li recovery processes.
[47]	Editorial Material	2019	Discussion of profitable recycling of low Co LIBs considering new process developments.
[48]	Editorial Material	2018	Study of currently used recycling strategies for LIBs.
[51]	Proceedings Paper	2019	The LCA of energy consumption and GHG from critical minerals recycling of LIBs.
[144]	Proceedings Paper	2015	The liquid membrane permeation with supported flat sheet membranes.
[31]	Article	2021	Up-to-date review on the methods for the recovery of Co.
[54]	Article	2021	Selective recovery of Co from nano-Co_3S_4 using PSL.
[55]	Article	2019	Co(II) extraction using toluene diluted Cyphos IL 102 and chemical precipitation; recovery Co, Li, Mn, Ni, Al, Fe, Cu.
[56]	Article	2010	Electrodeposition of Co, Cu multilayers.
[57]	Article	2020	Complexation-assisted solvent extraction of Co, Ni with factor 372; study of leaching kinetics Li, Ni, Co, Mn.
[58]	Article	2019	Carbothermal reduction in a muffle furnace, magnetic separation with fraction containing 90% of Co.
[59]	Article	2021	Electrochemical reduction of Co from $LiCoO_2$ that uses a molten salt fluidized cathode technique.
[60]	Article	2014	Hydrometallurgical process for recovery valuable metals from NCA, LIBS cathodes by using acids (H_2SO_4, HNO_3 and HCl); the recovery efficiency of Co (100%), Ni (99.99%).
[61]	Article	2021	The thermodynamic simulations for cobalt (II) hydroxide recovery; experimental; the precipitation under different pH conditions; optimal: pH 9, efficiency close to 100%.
[63]	Article	2014	Leaching and calcination of $Co(OH)_2$ and Co_3O_4 films with efficiency of 66.67% and 64.29%, respectively.

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Table A4. *Cont.*

Recovery of Materials			
Reference	**Type**	**Publication Year**	**Summary Content**
[64]	Article	2011	Electrodeposition of Co onto 430 steel in order to obtain Co_3O_4 film; recovery of pure Co using acidic dissolution of $LiCoO_2$.
[67]	Article	2017	Li recovery by ion sieve, two-step precipitation using Na_2CO_3 and Na_3PO_4; recovery rates 74.72% and 92.21%, respectively.
[68]	Article	2021	Metal-based strategy for selective Li leaching from NCM, LCO, and LMO by Co^{2+} or Mn^{2+}; 95% leaching rate, without metal ions left in the leachate; study of electrochemical performance of $LiCoO_2$ particles.
[69]	Article	2020	Oxalic acid-based recycling process; 90.13% purity of $LiCoO_2$.
[70]	Article	2016	Experimental study for $LiCoO_2$ recovery by using H_2SO_4 and HCl as leaching agents; optimal 2 M HCl, 60–80 °C, for 90 min; explanation of temperature influence.
[71]	Article	2018	Li recovery from low Li high-salt solution; precipitation of Li by P; the P/Li mass ratio of the catholyte reduced to 0.23 (the feed of 1.48), Li_2CO_3 precipitation rate reached 88.3% at 80 °C.
[72]	Article	2020	Recovery of $LiCoO_2$ via structure restoration; sintering of $LiCoO_2$ powder and Li salts mixture, improvement of regeneration using nanosized Al_2O_3 particles.
[73]	Article	2014	Ultrasonic-assisted leaching of Li, Co; testing H_2SO_4, HCL, and citric acid; optimal conditions for using 0.5 M citric acid with 0.55 M H_2O_2, a solid-to-liquid ratio of 25 g L^{-1}, a temperature of 60 °C, 5 h, and ultrasonic power of 90 W; recovery of 96% of Co and nearly 100% of Li.
[74]	Article	2021	Kinetic investigation of an oxalate-based process for recovery of Li and Co from $LiCoO_2$; a combined shrinking core model (cSCM) was used for $LiCoO_2$ digestions; description of importance of cost-effective, environmentally friendly, and energy-effective recycling processes.
[75]	Article	2020	Leaching valuable metals from $LiCoO_2$ powders using biodegradable organic MSA; leaching efficiencies of Li and Co are achieved at nearly ~100% and ~100%, respectively.
[76]	Article	2020	Hydrometallurgical-electro dialytic method for Li, and Co recovery from $LiCoO_2$; recovery rate 62% for Li and 33% for Co, whereas 80% of Co was electrodeposited at the cathode.
[77]	Article	2018	Recycling $LiCoO_2$ cathode powders using organic acids (CA, TA, AA) for metal recovery; almost complete dissolution of Li, nearly 90% dissolution of Co occurred at temperature of 80 °C after 6 h.
[78]	Article	2009	Recovering Co and Li using a combination of crushing, ultrasonic washing, acid leaching and precipitation; results: 97% of Li and 99% of Co was dissolved.
[79]	Article	2015	Thermodynamic (including thermogravimetry) analysis of possible reaction between $LiCoO_2$ and graphite; obtaining products of Co, Li_2Co_3 and graphite.
[80]	Article	2006	Recovery of $LiCoO_2$ including separation of Al foil using DMAC, and the PVDF and carbon powders elimination by calcining; elements morphology and structure analysis.

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Table A4. *Cont.*

Recovery of Materials			
Reference	**Type**	**Publication Year**	**Summary Content**
[81]	Article	2020	Suspension electrolysis system for directly recycling $LiCoO_2$ at atmospheric condition without any usage of acid and alkalis.
[83]	Article	2021	Laboratory-scale study of smelting reduction and recovery of nearly 100% recovery of Co, Ni, and Mn in the formed alloy and a nearly 100% recovery of Li in the flue dust.
[84]	Article	2021	Smelting reduction of Co, Ni, Mn (alloy), and Li (in the flue dust) in a pilot-scale Electric Arc Furnace demonstrated for two trials; recovery material yields over 90%.
[85]	Article	2018	A thermal treatment-ammoniacal leaching process; based on TG-DSC analysis calcination at 300 °C and 550 °C in air atmosphere. Complete leaching of Ni, Co, Mn, and Li with efficiencies of 98%, 81%, 92% and 98%, respectively.
[86]	Article	2019	A complex procedure of wet crushing, screening, and ternary leaching system composed of ammonia, ammonium sulfite, and ammonium bicarbonate for monitoring behavior of Li, Ni, Co, Cu, and Al. Almost fully leaching out of Ni and Cu, while Al is hardly leached, and Li (60.53%) and Co (80.99%) exhibit a moderate leaching efficiency.
[87]	Article	2020	A closed-loop recycling system of mixed organic acid leaching and sol-gel method resynthesis for $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ cathode material. Under optimal conditions (temp.: 70 °C, time: 30 min, slurry density: 20 g/L), the leaching efficiency of Li, Ni, Co, and Mn is 99.06%, 97.11%, 96.46%, and 97.22%, respectively.
[88]	Article	2020	Dissolution−chelation mechanism of the $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ cathode materials in acidic solution (malonic acid, hydrogen peroxide); under the optimal conditions the leaching efficiency of the Li is 95%, for Ni, Co, and Mn it reaches over 98%.
[89]	Article	2017	Leaching process for the recovery of Li, Ni, Co, and Mn from spent $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$-based LIBs and cathode scraps;the efficiencies for Li, Ni, Co, and Mn reached 99.7% under the optimized conditions.
[90]	Article	2020	A higher recycling ratio of valuable metals, that is, 94.9% of Li, 94.5% of Co, 94.4% of Ni, and 95.5% of Mn, was achieved by using sulfides and reducing the solubility of lithium carbonate in the lithium precipitation step at room temperature with ethanol.
[91]	Article	2020	Selective leaching system of NH_3–$(NH_4)_2CO_3$-Na_2SO_3 for NMC; single and multistage leaching; high recovery of 98.4% (Li), 99.4% (Co), 97.3% (Ni), and high-purity (>99%) $MnCO_3$ products were obtained.
[92]	Article	2019	Reduction-ammoniacal method (hydrothermal) devoted to the effects of various species of ammonia, ammonium salts, and reductants on the leaching of Li, Co, Ni, Mn, and Al from spent LIBs.
[93]	Article	2020	Process of physical separation of materials from spent LFP batteries; heat treatment, and corona electrostatic separation for metallic from the nonmetallic particles.

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Table A4. *Cont.*

Recovery of Materials			
Reference	**Type**	**Publication Year**	**Summary Content**
[94]	Article	2018	A hydrometallurgical process for recycling cathode materials dissolved in a mixed acid containing phosphoric and citric acid; leaching efficiency of ca. 100% for Li, 93.38% for Ni, 91.63% for Co, and 92.00% for Mn, respectively.
[95]	Article	2021	Regeneration process for a ternary cathode material using high-temperature calcination, and coprecipitation procedures; results: above 97.9% transition-metal elements and 81.2% Li element in the spent LIBs could be reused; NCM-r characterization using physical and electrochemical measurements compared to commercial NCM-c.
[96]	Article	2019	Approach for recovery of NMC particles while preserving their chemical and morphological properties, with a minimal use of chemicals.
[97]	Article	2017	Extraction and co-precipitation processes for 100% of Mn, 99% of Co and 85% of Ni recovery by D2EHPA in kerosene; Li recovery using the raffinate as Li_2CO_3 with the purity of 99.2% by precipitation method.
[98]	Article	2021	Recovery of valuable metals from $LiNi_{0.15}Mn_{0.15}Co_{0.70}O_2$ using the best leachant between HCl and H_2SO_4 + H_2O_2 resulting in almost 100% recovery.
[99]	Article	2016	Recovery by using D,L-malic acid; synthesis of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ through a sol-gel process.
[100]	Article	2020	Regeneration process for $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ using mixed acid leaching, oxalate co-precipitation and solid-phase reaction.
[101]	Article	2021	Froth flotation process for NMC111, and LMO materials separation; for multiple stages 95% grade or above of NMC111 in the froth product and 95% grade of LMO in the tailing product was separated.
[103]	Article	2022	Two-step leaching of the $LiNi_xCo_yMn_zO_2$ by sequential application of organic and mineral acids; ~99% of metals could be leached.
[102]	Article	2016	Regeneration of a $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ cathode material directly from the purified leaching solution via co-precipitation followed by solid-state synthesis.
[104]	Article	2019	Material recovery from NMC 811 using leaching by hydrochloric acid.
[105]	Article	2021	Three-step treatment for the separation of cathode components for sustainable LIBs recycling.
[106]	Article	2019	Two-step leaching of the exhausted $LiNi_xCo_yMn_zO_2$ by sequential application of both organic and mineral acids; achieving more than 99% efficiency for Li and Co recovery.
[107]	Article	2020	Eco-friendly recycling; the current collector of Al preforms as the in situ reductant of thermite reduction transforming valuable metals in $LiNi_xCo_yMn_zO_2$ cathode into $LiAlO_2$, Li_2O, NiO, CoO, and MnO.
[108]	Article	2016	A direct regeneration of cathode materials from spent $LiFePO_4$ batteries using a solid phase sintering.

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Table A4. *Cont.*

Recovery of Materials			
Reference	**Type**	**Publication Year**	**Summary Content**
[110]	Article	2017	A leaching process for recycling valuable metals using sol−gel method and lactic acid as a leaching and chelating agent; the leaching efficiency of Li, Ni, Co, and Mn reached 97.7, 98.2, 98.9, and 98.4%, respectively.
[109]	Article	2016	Three different separation processes, including direct calcination, solvent dissolution, and basic solution dissolution, were applied to obtain the active materials from LIBs scraps.
[111]	Article	2016	The leaching behavior of Ni, Mn, Co, Al, and Cu from treated cathode active materials; study of ammonium sulfite as a reductant, and ammonium carbonate as a pH buffer.
[112]	Article	2016	Process for recovery $LiNi_xMn_yCo_zO_2$ recovery.
[113]	Article	2015	Hydrometallurgical process using sulfuric acid leaching liquor (ammonium oxalate, saturated sodium carbonate solution) for treating waste cathode materials; recovery efficiencies attained as follows: 98.7% for Ni; 97.1% for Mn, 98.2% for Co and 81.0% for Li under optimized experimental conditions.
[114]	Article	2012	Recycling NiMH and LIBs using three mechanical treatment routes for each type followed by chemical leaching.
[119]	Article	2020	Spent carbon cathode recycled (kind of roasting technologies) and used applied as the anode of Li-ion batteries (LIBs).
[120]	Article	2019	Strategy devoted to preparation of Si/CNF/C composite for LIBs by using self-prepared micron-sized silicon and waste high-density polyethylene (HDPE) as raw materials.
[178]	Article	2019	Preparation of carbon paper that is coated with recycled silicon powder (CP-RSP); it acts as both the current collector and the active material for the anodes of LIBs.
[121]	Article	2020	Analysis of reusing spent graphite as anode material for LIBs and SIBs after reconstruction process.
[122]	Article	2021	Preparation of ORR electrocatalyst applied in fuel cells based on recycled anode graphite of spent LIBs.
[125]	Article	2021	Lab-scale DN50 pulsed disc and doughnut column solvent extraction of Mn from LIBs resulting in a 94% extraction yield.
[127]	Article	2021	Extensive characterization analysis (including XRD, EPMA, XANES, etc.) of slags of the system Li_2O-CaO-SiO_2-Al_2O_3-MgO-MnO_x with up to 17 mol% MnO_2 content for recycling.
[128]	Article	2020	Direct recycling of cathode scrap from spent LIBs based on sulfate radical-based advance oxidation processes (SR-AOPs) that entail a complex synthesis process.
[129]	Article	2016	An environmentally friendly Mn-based material, where binder-free self-supporting (BFSS) electrodes are prepared using a fibrous, high aspect ratio MnO_2 active material.
[130]	Article	2015	A green process route for recycling $LiFePO_4$/C materials using a crystalline $FePO_4 \cdot 2H_2O$ phase (metastrengite I).
[131]	Article	2015	An approach for reuse, recycle, and regeneration of a spent LFP cathode for rechargeable lithium- and sodium-ion batteries.

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Table A4. *Cont.*

Recovery of Materials			
Reference	**Type**	**Publication Year**	**Summary Content**
[132]	Article	2017	Direct regeneration for scrapped LFP; high yield of high-purity products of cathode material mixture ($LiFePO_4$ + acetylene black), anode material mixture (graphite + acetylene black) and other outputs (shell, Al foil, Cu foil, and electrolyte solvent, etc.).
[147]	Article	2016	Conversion method for the recycled LCO from spent LIBs into an efficient electrocatalyst for oxygen evolution reaction (OER); after 500 cycles a current density of 9.68 mA cm^{-2} at 1.65 V.
[138]	Article	2019	A high voltage/energy and long life $LiFe_{0.6}Mn_{0.4}PO_4/C$ (LFMP/C) composite prepared by reusing the whole $LiMn_2O_4$ cathode.
[139]	Article	2019	The soft-chemical treatment non-destructively recycles cathodes: NCM 523 and NCM 622; the reproduction of electrodes with performance equivalent to the original.
[140]	Article	2018	Four representative recycling streams were produced by a hydroxide co-precipitation to demonstrate the flexibility of the recycling process and generation of consistentquality cathode materials (NMC111).

Table A5. Assessed literature in review for "Recycling of Materials" category.

Recycling of Materials			
[32]	Review	2020	Recycling strategies for valuable metals in mixed-metal LIB cathodes and scrap for different types of chemistries.
[33]	Review	2020	Advances in the anode and cathode materials for the next-generation LIBs.
[133]	Article	2019	Mechanical separation and high-temperature pyrolysis of used LFP cathode active materials from retired EVs.
[134]	Article	2021	The SC CO_2 extraction of organic binders from spent LIBs to facilitate the liberation of the cathode material from Al foil.
[135]	Article	2015	Thermal decomposition of the PVDF binder used between coating and foil; ANVIIL separation process.
[136]	Article	2021	Complete separation of positive active materials from Al foil, without foil destruction.
[137]	Article	2020	In situ separation and recycling procedure of coating materials and Al foils from spent LIBs using ultrasonic-assisted acid scrubbing method.
[141]	Article	2020	High-capacity Si/C anode LIBs materials that are based on Si and lignin waste from PV and the traditional paper industry.
[142]	Article	2017	Recycling procedure for waste Cu scraps in the form of CuCl powders via the facile hydrothermal route.
[143]	Article	2020	Adsorption performance of spent LFP and LMO cathodes as adsorbents toward heavy metals in water.
[145]	Article	2020	The pyrolysis kinetics of active cathode material using various methods, including Flynn–Wall–Ozawa (FWO), Friedman, Kissinger–Akahira–Sunose, Starink, Tang, and Boswell.

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Table A5. *Cont.*

Recycling of Materials			
[146]	Article	2019	A study focused on using collected silicon oxides (SiOx) particles that are condensed from Si vapors exhausted from the ingot-growing furnace as an anode material.
[8]	Article	2016	Summary of procedures for the recycling and recovery of spent LIBs.
[7]	Article	2018	Review of the state-of-the-art techniques for metal recycling from spent LIBs.
[46]	Article	2020	Summary of technologies and issues in the disposal of spent LIBs from EVs.
[124]	Article	2015	Separation of the active electrode materials from the Co and Al foils in case of post-vehicle-application LIBs.
[123]	Article	2020	Separation of electrode materials from the current collectors using ethylene glycol.
[52]	Article	2020	Importance of recovering critical materials and improving battery designs from the cell to module level to facilitate recyclability.
[53]	Article	2013	Insulation of minerals processing operations and their effects in the case of LIBs and NiMH scraps.
[34]	Article	2020	The perspectives of hydrometallurgy and pyrometallurgy, including the process of optimization and novel recycling.
[50]	Article	2021	Global warming potential of a new waterjet-based recycling process for cathode materials of LIBs.
[49]	Article	2018	Overview of challenges in the material supply chain for automotive LIBs.

Table A6. Assessed literature in review for "Economical Evaluation" category.

Economic Point of View			
[10]	Review	2019	An analysis of recycling technologies from a CE perspective; overview of currently-in-service recycling facilities.
[167]	Review	2019	A circular economy insight devoted to energy storage systems on the world-wide scale.
[168]	Review	2020	Study of a circular economy for LIBs recycling techniques; a literature review of opportunities and challenges for recycling LIBs; recycling patents.
[169]	Review	2021	Environmental and economic aspects of recycling by hydrometallurgical processes.
[170]	Proceedings Paper	2012	Review of the state-of-the-art of recycling processes of spent LIBs based on the $LiCoO_2$ system.
[171]	Proceedings Paper	2015	Different scenarios for the return rates of LIBs from EVs considering their first and second life application.
[172]	Proceedings Paper	2019	Insights for future development options for recycling EOL LIBs based on the techno-economic analyses.
[173]	Proceedings Paper	2018	The economic profits estimation of recycling spent LFP and NMC batteries in conditions of China.

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Table A6. *Cont.*

Economic Point of View			
[174]	Article	2019	An AI approach for evaluating the residual energy of the LIBs embedded in battery packs used in EVs.
[4]	Article	2021	The techno-economic model for comparing recycling locations and techniques; six different locations, five LIB types.
[175]	Article	2013	Optimization model for the profitability analysis for recycling facilities of LIB technologies.
[176]	Article	2017	Leaching of NMC LIBs using acetic and maleic acid with 98% recovery of valuable metals; the economic analysis of performed hydrometallurgical technique.
[177]	Article	2020	An environmental economic model based on a real case study from a Chinese EV manufacturer.

Table A7. Assessed literature in review for "Recycling of EV LIBs" category.

Recycling of EV LIBs			
[36]	Proceedings Paper	2016	Hazards and consequences of incorrect recycling processing and procedures.
[37]	Proceedings Paper	2013	Overview of the framework of EV LIBs recycling.
[35]	Article	2020	Solutions for the screening and regrouping of retired LIBs considering the secondary application and future recycling.
[179]	Correction	2020	Correction to review article by Harper et al. [180].

Table A8. Assessed literature in review for "Recycling of LIBs" category.

Recycling of LIBs			
[38]	Review	2020	Critical overview devoted to sustainability of LIBs recycling processes.
[39]	Review	2020	A guide for suited recycling methods for metal recovery and future repurposing of spent LIBs.
[40]	Review	2020	Categorization according to state-of-the-art schemes of waste treatment technology in terms of LIBs recycling.
[41]	Review	2018	Review devoted to recent advancements in recycling technologies of spent LIBs.
[42]	Review	2020	A comparison of recycling challenges, processes, and impacts in the case of ASSBs and LIBs.
[115]	Proceedings Paper	2010	LCO batteries' recycling based on sulfuric and oxalic acid.
[148]	Proceedings Paper	2017	Secondary Al production by spent LIBs' recycling.
[149]	Proceedings Paper	2014	Lithium recovery from seawater by electrodialysis.
[82]	Article	2019	Ammonia leaching recycling procedure for LIBs.
[116]	Article	2007	Two recycling processes for spent LIBs: calcination and fusion.
[150]	Article	2016	Waste-to-Li system based on electrochemical reaction with water and Li precursors.
[151]	Article	2019	Critical discussion of Polish waste treatment management systems and disposal solutions.

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Table A8. *Cont.*

Recycling of LIBs			
[152]	Article	2019	Recycling procedure for spent LIBs; comparison of recycled materials with LNCM-R and LNCM-N.
[153]	Article	2014	Eco-friendly process for Li recycling from organic electrode materials for secondary used LIBs.
[154]	Article	2013	Chemical analysis, questionnaire survey, and flow analysis for Co recovery from LIBs recycling in conditions of Japan.
[155]	Article	2019	Direct physical and combined recycling procedure for LIBs.
[156]	Article	2021	LIBs discharging in aqueous salt solutions; performance and optimalization.

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ORIGINAL PAPER

Lithium-ion (LCO/NMC, NMC, LFP) battery recycling: partial LCA study

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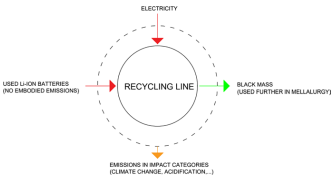
Abstract
Recycling lithium-ion batteries (LIBs) have become increasingly important in response to expanding electromobility. This paper is focused on evaluating the environmental impacts (EIs) of recycling pre-treatment of three types of LIBs with black mass as its product. A detailed gate-to-gate Life Cycle Assessment study was conducted to obtain EIs of the recycling process. The benefits of LIBs recycling pre-treatment and significant recovery of secondary aluminum for compared battery types are highlighted in the analysis. This paper points out that the varying chemistry of the compared LIBs does not affect the resulting EIs of the recycling pre-treatment procedures.

Graphical abstract



Keywords Material science · Ecology · Metals · Electrotechnology · Battery · Recycling

Introduction

The expansion of electromobility, especially in the automotive industry, leads to an increasing number of waste electric vehicles and their battery systems. Thus, the necessity of end-of-life (EOL) lithium-ion batteries (LIBs) treatment is set down; valuable materials such as cobalt, nickel, manganese, and lithium, and other essential metals such as aluminum, copper, or iron can be obtained from the recycling of waste batteries.

Reusing secondary materials in new products mitigates the environmental impacts of the sectors within the vehicle industry supply chain. That means, for example, less emission from mining ores and minerals and less energy demand to produce primary materials used in batteries.

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This paper presents the partial gate-to-gate Life Cycle Assessment (LCA) analysis focused on the LIBs recycling pre-treatment evaluation. The environmental impacts (EIs) of recycling three different types of LIBs: lithium cobalt oxide/lithium nickel manganese cobalt oxide (LCO/NMC), lithium nickel manganese cobalt oxide (NMC), and lithium iron phosphate (LFP) were compared. Each battery type contains different inner materials: cathode active materials, electrolyte, separator, cathode aluminum foil, anode copper foil, anode active materials, and housing [1]. The battery recycling pre-treatment process included the following steps: discharging, dismantling, crushing, milling, sieve separation, magnetic separation, and a high-temperature furnace for thermal treatment [2].

The performed study focuses on the recycling pre-treatment procedure, considering black mass as the final output under the conditions of the plant located in the Czech Republic. Black mass (BM) represents shredded materials with a significant valuable metal content (such as cobalt,

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Table 1 Environmental impacts of battery recycling

Impact category	Unit	LCO/NMC	NMC	LFP
Climate change	kg CO_2 eq	– 12.882	– 12.861	– 12.859
Particulate matter	Disease inc	– 0.0014	– 0.0014	– 0.0014
Acidification	mol H^+ eq	– 168.588	– 168.503	– 168.494
Resource use, fossils	MJ	– 136.205	– 135.869	– 135.832

manganese, or nickel); it is typically used for further specialized metal recovery based on pyro-/hydro-metallurgy recycling methods [3]. This paper points out that the varying chemistry of the compared LIBs does not affect the resulting EIs of the recycling pre-treatment procedures.

Results and discussion

The functional unit of the LCA study is one ton of produced black mass (BM). To this referring amount, the EIs were counted. In the normalization and weighing phase of the LCA study, the most important impact categories of the Environmental Footprint 3.0 (EF 3.0) method were named. The significant normalized impacts of the recycling process, summarized in Table 1, are in EF 3.0 impact categories:

- Climate change (kg CO_2 eq);
- Acidification (disease inc.);
- Particulate matter (mol H^+ eq);
- Resource use—fossil (MJ).

The total EIs are negative, which signifies the battery recycling benefits. Although the composition of LIBs is different for each battery type, the EIs of their recycling route are nearly equal. There is a maximum of 0.3% nuance between the EIs of each battery type. It comes from the different compositions of each battery type and generated shares of black mass and secondary aluminum. The chemical composition of selected LIBs has a negligible effect on EIs considering recycling pre-treatment.

However, the main advantages of battery recycling are set for obtaining secondary aluminum as it goes with nearly 96% of total EIs. This is due to that recycled aluminum can save up to 95% of energy compared to primary aluminum. The rest comes from obtaining secondary graphite and steel. The production of primary aluminum is highly demanding on electrical energy and ores. Electricity used in the production stage comes in the Czech Republic mainly from fossil fuels [4]. In a nutshell, recovery of secondary aluminum and its further usage can decrease the EIs of this heavy industry.

Conclusion

According to the performed study, the crucial benefit of recycling pre-treatment of different LIBs was determined in gaining secondary aluminum. The main advantage is preventing primary aluminum production, which is highly demanding on energy and metal ores. The negative EIs coming from obtaining secondary aluminum denote the benefits of the recycling stage of LIBs.

This paper points out that the varying chemistry of the compared LIBs does not affect the resulting EIs of the recycling pre-treatment procedures. The follow-up work

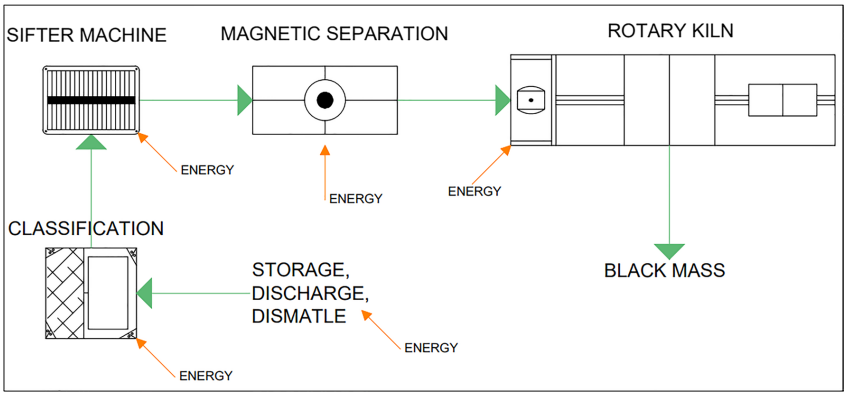

Fig. 1 Recycling pre-treatment process scheme

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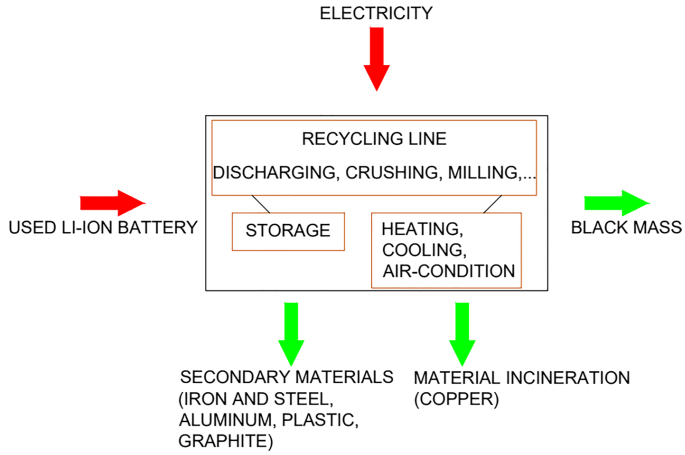

Fig. 2 LCA boundaries and material flows

will focus on the full LCA study comparing recycling pre-treatment routes of consumer electronic batteries to electric vehicle battery packs.

Experimental

The battery recycling pre-treatment process, outlined in Fig. 1, included the following steps: discharging, dismantling, crushing, milling, sieve separation, magnetic separation, and a high-temperature furnace for thermal treatment

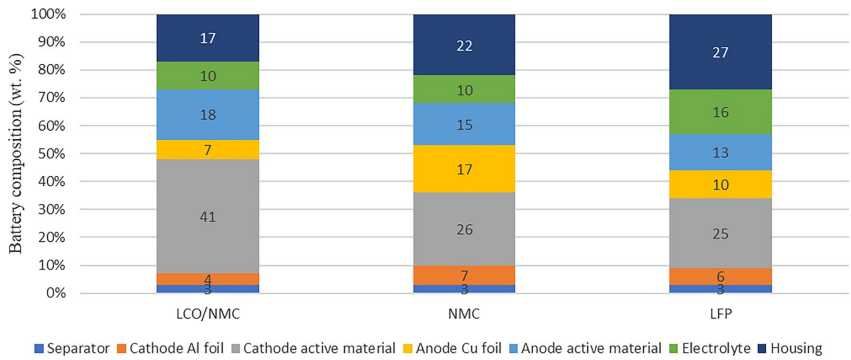

Fig. 3 Material composition of studied battery types

Table 2 Material inventory of recycling pre-treatment process

(In tonnes)	Milling	Sifter machine	Magnetic separator		Rotary Kiln		
Compound	Plastic	Iron/steel	Cu	Al	Electrolyte	Graphite	BM
Treatment	R	R	I	R	I	R	–
LCO/NMC	15.75	89.25	36.75	21.00	52.50	94.50	215.15
NMC	15.75	115.50	89.25	36.75	52.50	78.25	136.50
LFP	15.75	141.75	52.50	31.50	84.00	68.25	131.25

R Recycling, *I* incineration, *BM* black mass

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[2]. The evaluated EIs, by a detailed gate-to-gate LCA study, were examined within systems boundaries illustrated in Fig. 2.

Three different types of LIBs for recycling were compared in this work. An annual battery mass of each battery type is considered 525 tons. This refers to the exemplary recycling plant already in operation but with reduced operating shifts [5]. Their material shares (within anode/cathode active materials, electrolyte, separator, copper (Cu)/aluminum (Al) foils, and housing) are specified in detail in Fig. 3 [1] and are used for computation of material inventory in Table 2. As an input, the used LIBs are considered to be with zero embodied emissions.

A detailed gate-to-gate LCA study was conducted via 9.5.0.0 version of SimaPro modeling software using the Ecoinvent 3.9. database with a Cut-off (U) modeling access. The results were evaluated by Environmental Footprint 3.0 (EF 3.0) life cycle impact assessment method.

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Data availability The core data for this study were sourced from the licensed Ecoinvent 3.9 database and were transformed based on information from references [1–5].

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Gate-to-gate life cycle assessment of lithium-ion battery recycling pre-treatment

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ABSTRACT

Recycling spent lithium-ion batteries (LIBs) is critical for enhancing environmental sustainability and resource conservation; however, the environmental and energy impacts of LIB recycling are not yet comprehensively understood due to the diverse applications of LIB cells and the variability in recycling technologies. This study presents a gate-to-gate life cycle assessment (LCA) of a recycling pre-treatment process at a small-scale plant in the Czech Republic, focusing on spent LIBs from electric vehicles (EVs) and consumer electronics cells (CECs). Using the SimaPro LCA software and the Ecoinvent 3.9 database, the analysis evaluated the environmental impact of recycling operations across several categories, including climate change, eutrophication, freshwater, and resource use, minerals and metals. The findings reveal that the recycling pre-treatment process for CECs achieves greater benefits in climate change mitigation compared to EV batteries, with a 5% lower impact for climate change associated with EV batteries relative to CECs. Moreover, the study highlights the effectiveness of optimized recycling practices in alleviating environmental burdens. A notable finding is the significance of secondary material recovery, particularly metals such as copper and aluminium, as these materials can substitute for primary raw materials, thereby minimizing resource use and reducing emissions. These aspects emphasize the need for high recovery efficiency to enhance environmental benefits. However, further research is essential to fully comprehend the environmental impacts of LIB recycling and to resolve uncertainties concerning battery composition and the effectiveness of different recycling technologies.

1. Introduction

Lithium-ion batteries (LIBs) have become indispensable in modern technology due to their high energy density, lightweight design, and rechargeability. They power various devices, from portable consumer electronics to light or heavy means of transport, such as electric vehicles (EVs) or renewable energy storage systems (Kumar et al., 2023; Nitta et al., 2015; Sharma et al., 2020). The LIBs integration within transportation leads to a revolution in the automotive sector by providing a cleaner and more sustainable alternative to traditional internal combustion engines (Reddy et al., 2024; Ren et al., 2023). Vehicles equipped with LIBs offer reduced greenhouse gas emissions (GHGs) and contribute to mitigating climate change (Jannesar Niri et al., 2024; Reddy et al., 2024). Nevertheless, the environmental benefit should be considered within the entire lifespan of the product, process, or activity (Ilgin and Gupta, 2010). This includes not only the manufacturing and use phases but also the disposal or end-of-life (EOL) stage (Klöpffer and Grahl, 2014).

Recycling holds significant benefits for both the environment and the economy (Kwade and Diekmann, 2018; Pražanová et al., 2022a, 2022b). Commonly used LIBs are rich in valuable metals in battery electrodes, such as cobalt, lithium, manganese, and nickel, and other essential metals and minerals, including aluminium, copper, graphite, or iron within the rest of battery components, such as battery collectors and casings (Ferg et al., 2019; Li et al., 2018; Pražanová et al., 2022a). Considering the still-rising demand within a wide range of their application fields, recycling LIBs is crucial for effective waste management. Moreover, recycling conservates virgin material sources, reduces the environmental impact associated with mining and extraction, such as the emissions of GHGs, and contributes to the circular economy by closing the loop on selected materials (Mossali et al., 2020; Zheng et al., 2018). However, even the recycling processes are energy-intensive, generate emissions into water and air, and pose the risk of releasing toxic substances into the environment (Pražanová et al., 2022b; Zhang

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et al., 2018).

Life cycle assessment (LCA) is typically used to thoroughly quantify the environmental impacts of a product. LCA should ideally cover all stages of the product's life cycle, from raw material extraction to disposal ("cradle-to-grave"), but it can also focus on specific life cycle stages ("gate-to-gate") (Klöpffer and Grahl, 2014). Evaluating specific life cycle phases is essential for understanding the complexity and impact of procedures. This assessment enables the identification of opportunities for further optimization, ultimately reducing the overall environmental impact of the monitored processes (Ilgin and Gupta, 2010; Klöpffer and Grahl, 2014).

Very few LCAs addressing LIB recycling treatment have been documented in published literature. Moreover, the works vary significantly in their aims, scope, level of detail, assumed battery type, composition chemistry, and the employed assessment method. A comprehensive analysis of the impacts of recycling using cradle-to-gate LCA, focusing on energy consumption and greenhouse gas emissions of automotive LIBs, was provided by Dunn et al. (2012). Their work covered the "closed loop" recycling by pyrometallurgy, hydrometallurgy, and direct physical separation of the lithium manganese oxide (LMO) cathode and current collector materials. According to their approach, the direct physical recycling of LMO, aluminium, and copper could reduce energy consumption during material production by up to 48%. A similar study was provided by Hendrickson et al. (2015), who dealt with closed-loop pyrometallurgical and hydrometallurgical processing of LMO. Their finding revealed that material recovery from pyrometallurgy can mitigate environmental impacts associated with LIB production via a 6–56% reduction in primary energy demand and a 23% reduction in GHG emissions compared to virgin production. Further, the assessment of Elwert et al. (2015) focused on hydrometallurgical recycling of nickel manganese cobalt oxide (NMC) LIBs, highlighted the environmental benefits resulting from the recovery of the outer steel casing to a lesser degree, the cobalt and nickel present in the cathode materials. Rajaeifar et al. (2021) presented a comparative study of different pyrometallurgical techniques. In contrast, Cusenza et al., 2019 reported an evaluation of recycling an LMO-NMC LIB pack using a combination of pyrometallurgical and hydrometallurgical processes.

Despite the progress made, there is still a significant knowledge gap in the environmental assessment of the initial stage of recycling LIBs, referred to as recycling pre-treatment (Chan et al., 2020; Fujita et al., 2021; Pražanová et al., 2022a). This phase includes battery discharging, disassembly, crushing, rough material separation, and the removal of organic components through thermal treatment. The result of this process, known as black mass, is a finely ground mixture composed of various materials extracted from battery components, notably containing valuable metals (Kwade Arno & Diekmann Jan 2018). A recent study by Fridrich et al. (2024) provides a detailed gate-to-gate LCA of the recycling pre-treatment process for different types of LIBs, emphasizing the environmental impacts and significant recovery of secondary aluminium, and finding that variations in battery chemistry do not significantly affect the environmental impacts of these procedures.

This paper addresses a critical gap in the current understanding of LIB recycling by focusing on the pre-treatment phase, which has received limited attention in the literature. While most studies analyse the environmental impacts of the complete recycling process, the pre-treatment stage, which is crucial for material recovery and downstream process efficiency, has been underexplored. With the increasing adoption of LIBs, particularly in EVs and CECs, the demand for efficient and environmentally responsible recycling processes is more pressing than ever.

This study offers a detailed gate-to-gate LCA of pretreatment process in the case of a small-scale recycling plant in the Czech Republic, specifically examining the pre-treatment of LIBs from EVs and CECs. By analysing key environmental impact categories, such as climate change, eutrophication, freshwater use, and resource depletion, the paper highlights the considerable environmental benefits of optimizing the pre-treatment process. To reflect market realities, the study selected representative battery types from both EVs and CECs. For EV batteries, the focus was on lithium nickel manganese cobalt oxide (NMC), a leading chemistry in modern electric vehicles. For CECs, the analysis included lithium manganese oxide (LMO), lithium iron phosphate (LFP), and lithium nickel cobalt aluminium oxide (NCA) in equal proportions, capturing the diversity of battery chemistries used in consumer electronics. Given that pre-treatment is a necessary step in LIB recycling, as it enables the efficient recovery of valuable materials while meeting regulatory constraints (EUR-Lex, 2023; United States Environmental Protection Agency, 1976), a scenario without pre-treatment was not considered in this study. Alternative approaches, such as landfilling, are legally restricted in many regions (EUR-Lex, 2023), and direct incineration is not viable due to the economic value of recoverable metals like cobalt and nickel (Li et al., 2018). Consequently, this research focuses on evaluating and improving pre-treatment processes as they represent the only practical and sustainable pathway for EOL LIB management. The results show that recycling CECs yields a more substantial reduction in climate change impacts compared to the recycling of EV batteries. Additionally, the study identifies opportunities to improve resource efficiency and reduce emissions by enhancing pre-treatment practices, especially in small-scale facilities that serve as critical components of the LIB recycling network. At the same time, the recovery of critical metals like copper and aluminium plays a pivotal role in minimizing resource use and emissions. This research offers valuable insights into the environmental performance of LIB recycling and highlights the critical role of achieving high recovery efficiency to fully unlock these technologies' sustainability potential.

By refining existing methods and expanding the scope to include various battery types, the study contributes to the ongoing efforts to optimize recycling processes in response to growing global battery demand. Furthermore, the findings bridge a significant gap in the existing literature by focusing on the environmental performance of pre-treatment, offering a basis for future comparative studies and advancing the understanding of this critical stage in LIB recycling.

2. Materials and methods

2.1. Goal, scope, and assumptions

LCA is a standardized method used to assess the environmental impacts of a product, process, or service throughout its life cycle, from raw material extraction to disposal (cradle-to-grave). Partial LCA studies, or "gate-to-gate" analyses, focus on specific life cycle stages to evaluate their impacts. This study performed a gate-to-gate LCA to assess the environmental impacts of the pre-treatment phase in LIB recycling (Klöpffer and Grahl, 2014). The system boundary of this study is defined as encompassing the recycling pre-treatment processes of EOL LIBs from EVs and consumer CECs, covering five main steps: deep discharging, disassembly, crushing, separation, and thermal treatment. Inputs include EOL batteries, energy, and auxiliary materials, while outputs include recovered black mass, secondary materials (e.g., copper and aluminium), lost fractions, and hazardous waste such as wastewater. The detailed system boundary is illustrated in Fig. 1 (c). Moreover, it is assumed that using recycled materials avoids the environmental impacts associated with producing new materials, thus reflecting the benefits of avoided products. The functional unit for this assessment is defined as one metric tonne of recycled batteries, representing the amount of material processed through the pre-treatment phase. This choice ensures consistency with LCA best practices and facilitates direct comparison with similar studies in the literature. The selection of one metric tonne as the functional unit also aligns with industry norms and reflects a practical scale for evaluating the environmental performance of small-scale recycling facilities. Additionally, this approach captures the diversity of battery chemistries processed and their respective contributions to environmental impacts, as detailed in the system boundaries.

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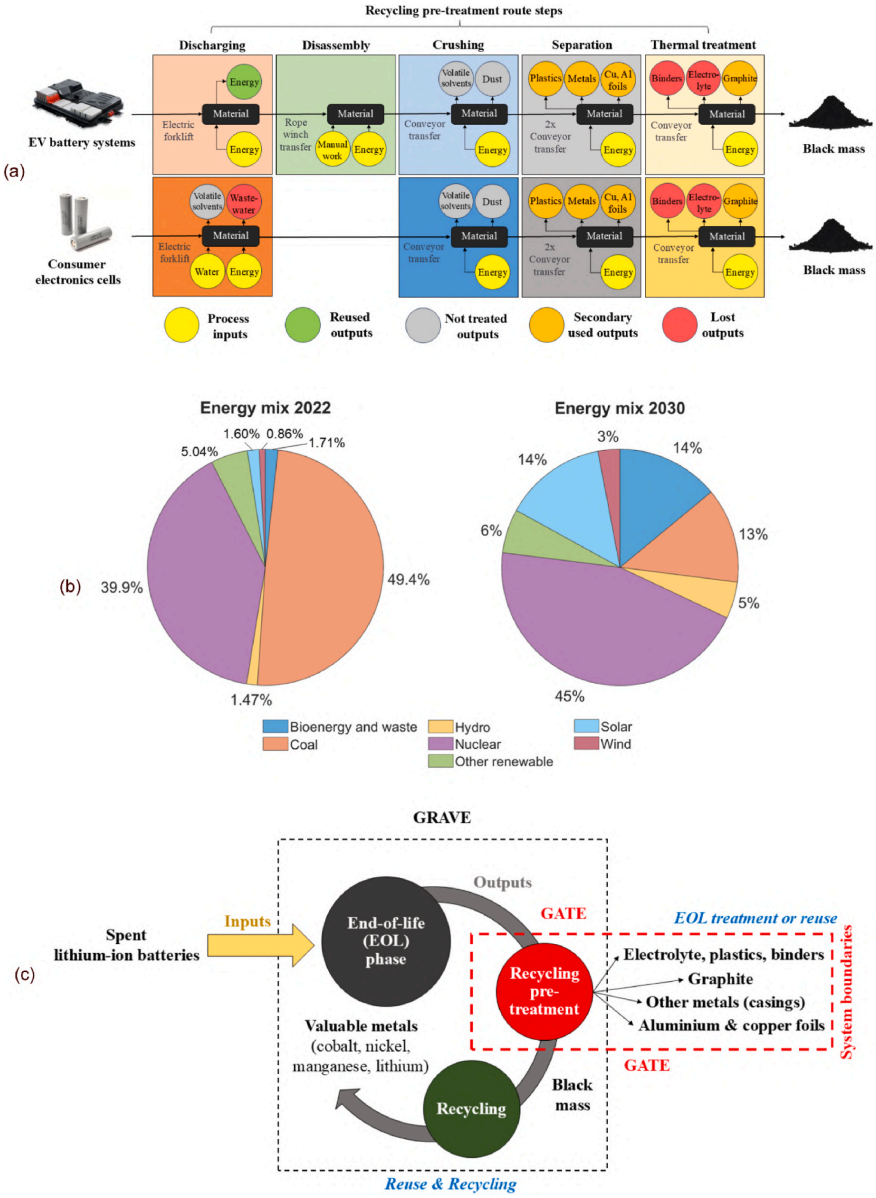

Fig. 1. (a) Process scheme of the recycling pre-treatment steps for spent lithium-ion batteries from electric vehicles and consumer electronics; (b) energy mix composition for analysed scenarios in 2022 and 2030 (CEPS, 2021); (c) end-of-life (EOL) phase of lithium-ion battery life cycle with a system boundaries highlighted in red, related to gate-to-gate life cycle analysis of recycling pre-treatment.

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The study presents a case study of a recycling plant in central Europe, specifically in the Czech Republic, which is dedicated to the recycling pre-treatment of spent LIB systems from EVs and consumer CECs. This evaluation is focused on study case scenarios grounded in established best practices, extensive experience in the field, relevant literature, and informed estimates based on existing research and data. The scope explicitly excludes downstream processes, such as refining of recovered black mass or reuse of electrolyte residues. A schematic illustration of the five-step recycling pre-treatment process studied in this work, including the characterized process inputs (EOL batteries) and outputs, such as reused, not treated, secondary and lost materials, is presented in Fig. 1 (a). Fig. 1 (c) provides a detailed depiction of the system boundaries, clearly delineating the cradle-to-gate processes included in this analysis. Detailed information on consumption and individual quantities is provided in Supplementary Materials (SM).

The whole process starts with a deep discharge step, which is essential for further safe and promising processing (Liu et al., 2022; Pražanová et al., 2022a). For EV technology, the electric active discharging procedures are typically preferred (Kwade Arno & Diekmann Jan 2018). Thus, for the case of EV battery systems (packs), a suitable battery discharge system with reduced energy consumption and high efficiency with a recovery energy capability of up to 96% is used (Keysight, 2024). The discharge process is included within the system boundary and contributes to annual energy consumption of 19.2 MWh. Within the CECs treatment, electrochemical discharge in 10 wt % sodium chloride (NaCl) and water solution, as the favourable discharging procedure, is applied (Ojanen et al., 2018; Rouhi et al., 2021). Discharging is conducted under strict safety protocols, including the ventilation of toxic fumes and continuous monitoring (Kong et al., 2018; Work Australia, 2012). The discharge bath, with a total volume of 2.5 m^3, is filtered as part of the operation and reused throughout the year.

The battery packs are manually disassembled using only small power tools at the battery module level due to the high complexity and wide variety of treated systems (An, 2019). This disassembly stage is part of the defined system boundary, with manual labour and power tools representing the primary resource consumption. The next step involves crushing either battery cells or entire battery units, facilitating the separation of their materials (Kwade Arno & Diekmann Jan 2018; Pražanová et al., 2022a). The modules are subsequently crushed in a two-step procedure using a double-shaft rubber recycling crusher and a rotor impact mill (Kwade Arno & Diekmann Jan 2018). The system boundary includes emissions of volatile solvents or particulate matter during this stage (Kwade Arno & Diekmann Jan 2018), which are captured by ventilation systems but not further processed within this analysis. The scope of this study does not cover their more detailed processing or reusing. Crushing CECs is done likewise. During the electrochemical discharging, volatile solvents and electrolyte are released into the solution (Pražanová et al., 2023). The subsequent disposal of this wastewater as hazardous waste (He et al., 2024; Mondal et al., 2024), along with its energy and resource demands, falls within the system boundary. The treated cells are rinsed with water and crushed using the same process steps as in the EV route. The final product of this line is black mass, i.e., a finely ground mix of fractions of battery components (Kwade Arno & Diekmann Jan 2018), which has been treated with selected processes (separation and thermal treatment) to remove plastics, binders, conductive additives, electrolyte residues, or other materials such as current collector metals (copper and aluminium), graphite from anode layers or casing materials (iron and steel). Outputs from this stage, such as black mass, metals, and other recyclable fractions, represent the final outputs within the system boundary. The study does not include further downstream processing of these outputs. More detailed information on the individual processes can be found within the SM.

The assessment was conducted for a case study of a small-scaled pilot recycling line, explicitly defining the system boundary to focus on pre-treatment operations, including discharging, disassembly, and crushing processes, but excluding downstream processing steps such as hydrometallurgical or pyrometallurgical recycling. The pilot line handles approximately one-third of the processing capacity compared to the current operations in central Europe (Baum et al., 2022). Thus, a single-shift operation (8 working hours) for 252 days per year, reflecting realistic conditions for small-scale facilities, was considered. The processing of EV battery systems and CECs in the recycling plant was assumed to occur in separate production cycles and is not done simultaneously. For clarity, the operational schedule was divided as follows: EV battery packs are processed for 240 days annually, while CECs are processed once a month for 12 days.

For EV battery systems, each battery pack was assumed to weigh between 400 and 420 kg, with 5 systems processed per shift, resulting in a total annual processing capacity of 500 tonnes. For CECs, a different processing approach was applied: 4 tonnes of batteries per shift were processed, resulting in a total annual capacity of 48 tonnes. To better contextualize the study, the EV battery systems processed in the pilot line were assumed to predominantly consist of lithium nickel manganese cobalt oxide (NMC) technology. This reflects one of the leading chemical compositions of EV LIBs today. In contrast, the CEC processing line was considered to handle batteries based on lithium manganese oxide (LMO), lithium iron phosphate (LFP), and lithium nickel cobalt aluminium oxide (NCA), all in equal proportions (16 tonnes of each type annually). Material proportions and the mass of individual battery components were detailed to enhance traceability and provided in Table 1 for reference.

In the assessed case, the recycling hall of the plant spans an area of 200 m^2, complemented by a 100 m^2 battery warehouse. The evaluation encompasses energy consumption for heating, air conditioning, security monitoring, and additional ventilation during electrochemical discharge or battery unit crushing. The SM provide detailed information regarding the energy consumption of individual recycling preparation or revision processes, totalling 0.18 GWh/year of plant operation.

2.2. Evaluated scenario and sensitivity analysis

This study evaluates the plant's operation dedicated to recycling pre-treatment of spent LIBs in their EV battery packs, and CECs type was conducted based on an implementation scenario (Scenario 2022), which utilizes the average energy mix data of the Czech Republic for 2022. This year was selected due to data availability, as only complete data for 2022 were accessible when the study began in early 2023. Additionally, to illustrate the impact of changes in the energy mix composition over time, Scenario 2022 was compared to the predicted energy mix composition for 2030 (Scenario 2030). The year 2030 was chosen based on projections indicating a transition towards a greener energy mix, including a reduction in coal reliance and an increase in renewables such as solar power. The composition of the energy mix in 2022 relied on data from the Czech electricity and gas market operator (OTE) database (OTE, 2024). Predictions for the 2030 energy mix were drawn from the "Resource Adequacy Assessment of the Electrical Grid of the Czech Republic until 2040" (MAF CZ 2022) (ČEPS, 2021). The composition of considered energy mixes is shown in Fig. 1 (b).

Both case studies assumed ~100% efficiency for the recycling technology and a ~100% share of secondary use for the obtained materials. While these assumptions represent an idealized scenario based on optimistic projections, they may not fully reflect the recovery and separation rates achievable with existing technologies. The materials were separated with ~100% efficiency, ensuring the resulting black mass achieved the highest possible quality. This demonstrates a highly optimistic scenario, intended to evaluate the maximum potential environmental benefits of recycling. Such a scenario is valuable as it highlights the upper bound of potential environmental gains, providing a benchmark for future improvements and showing what can be accomplished under ideal conditions. However, it is important to note that real-world operations with lower efficiency may result in reduced

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Table 1
The material proportion and mass of treated lithium-ion battery components (Baazouzi et al., 2023; Deshwal et al., 2022; Pražanová et al., 2022a; Sobianowska-Turek et al., 2021). The mass of cathode active materials corresponds to the maximal amounts of possibly obtained black mass in its highest quality.

Cell type	EV cells		Consumer electronics cells (CECs)						
	NMC	Total (tonnes)	*LMO*		*LFP*		*NCA*		Total (tonnes)
Battery component	wt. %		wt. %	Total (tonnes)	wt. %	Total (tonnes)	wt. %	total (tonnes)	
Cathode	40	200	24	3.84	28	4.48	25	4.00	12.32
Al foils	15	75	17	2.72	19	3.04	15	2.40	8.16
Active materials	25	125	7	1.12	9	1.44	10	1.60	4.16
Anode	24	120	16	2.56	19	3.04	17	2.72	8.32
Cu foils	12	60	8	1.28	9	1.44	8	1.28	4.00
Graphite	12	60	8	1.28	10	1.6	9	1.44	4.32
Plastics	5	25	5	0.80	5	0.8	5	0.80	2.40
Metals	14	70	15	2.40	13	2.08	14	2.24	6.72
Binders	7	35	4	0.64	2	0.32	4	0.64	1.60
Electrolyte	10	50	36	5.76	33	5.28	35	5.60	16.64
Total	100	500	100	16	100	16	100	16	48

Note: EV – "Electric vehicle"; NMC – "Lithium nickel manganese cobalt oxide"; LMO – "Lithium manganese oxide"; LFP – "Lithium iron phosphate"; NCA – "Lithium nickel cobalt aluminium oxide".

environmental benefits compared to those presented in this study. Sensitivity analyses were conducted to account for these variations. Detailed quantities of the output materials are provided in Table 1. However, the limitation of this work is that the utilized data in Table 1 are based solely on previous literature, rather than directly collected from the field, due to either the unavailability of field data or their confidentiality. Furthermore, the potential for secondary use was maximized, aligning with the primary objective of recycling. In sensitivity analyses, materials not obtained as secondary materials (and whose environmental impacts are excluded from the system) represent the Cut-Off modelling approach.

The modelling approach defines avoided products as items prevented from coming into existence through specific interventions. In our system, secondary materials are treated as avoided products, often associated with negative environmental impacts, as their use offsets the production of new materials. This approach accounts for the environmental burdens these secondary materials help mitigate. Sensitivity analysis 1 (SA1) is dedicated to Scenario 2022 and describes the effects of the implemented technology efficiency, which directly impacts the quality of the obtained materials. The efficiency is influenced by the applied recycling methods and their ability to recover specific material categories, as listed in Table 2. For instance, the recovery of electrolyte is assumed to be 100%, while graphite recovery is considered 0% in variants A, B, and C. In variant D, the recovery efficiency of all materials is set to match the baseline Scenario 2022. These variants simulate the effect of technology improvements or degradations on recovery rates and highlight their influence on environmental outcomes. In this case, a ~100% secondary use of the recovered materials was assumed. The efficiency of the process is determined by the percentage of obtained materials, according to their materials type, including electrolyte, plastics, binders, graphite, other metals, and Al & Cu foils within the material flows of the recycling pre-treatment operation, as illustrated in Table 2. While a ~100% recovery rate was assumed for modelling purposes, real-world recovery rates for certain materials may be lower, which could affect the environmental benefit results. Detailed data are provided in SM.

Table 2
The assumptions used in the sensitivity analyses (SA) for evaluating the recycling pre-treatment scenarios of spent lithium-ion batteries.

Sensitivity analysis parameter	Material category	SA1 variants (based on Scenario 2022)				
		A	*B*	*C*	*D*	*Scenario 2022*
Technology efficiency	Electrolyte	100%	100%	100%	100%	100%
	Plastics, binders	0%				
	Graphite		0%			
	Other Metals	80%	80%	80%		
	Al & Cu foils				80%	

Sensitivity analysis parameter	Material category	SA2 variants (based on Scenario 2022)			
		X	*Y*	*Z*	*Scenario 2022*
Secondary usage	Electrolyte	0%	0%	50%	100%
	Plastics, binders				
	Graphite				
	Other Metals		100%		
	Al & Cu foils				

Sensitivity analysis 2 (SA2) is focused on assessing the secondary use effect in Scenario (2022). This analysis evaluates scenarios X, Y, and Z to explore the extent to which secondary material usage influences environmental outcomes. Variant X assumes no secondary use, meaning all materials are directed to landfills or incineration, while variant Y assumes partial secondary use (e.g., 50% for graphite). Variant Z assumes full secondary use of recovered materials, aligning with Scenario 2022. These variations test the dependency of environmental benefits on secondary material applications. The evaluation considers the highest quality black mass product (achieved through ~100% efficiency of the implemented technology), and the secondary usage shares for selected material groups are shown in Table 2.

In an ideal scenario, materials such as Al and copper Cu foils, metals, graphite, and plastics are considered secondary materials, which can be recycled and reused. However, electrolytes are typically only partially treated through wastewater or incinerated in kilns. Binders, on the other hand, are always directed to landfills. Furthermore, if secondary materials are not recycled, they end up in landfills. Within the provided sensitivity analysis, the scenarios are highly optimistic, demonstrating the maximum possible environmental benefits from recycling that can be achieved within the considered processes. Such an optimistic outlook underscores the potential for significant environmental improvements and serves as a guide for maximizing the efficiency of future recycling technologies. While the energy mix used to power the recycling pre-treatment line improves in time (emissions get lower), the environmental footprint of avoided products stays the same. This leads to a higher positive impact of battery recycling on the environment. The input data of this evaluation are given in SM.

2.3. Life cycle assessment approach

This study provides a partial gate-to-gate LCA for the selected

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recycling pre-treatment processes for spent LIBs, conducted via the 9.5.0.0 version of SimaPro LCA modeling software using the Ecoinvent 3.9. database with a Cut-Off approach and Environmental Footprint 3.1 (EF 3.1) life cycle impact assessment method (European Platform on LCA | EPLCA, 2024). The system boundaries focus on the pre-treatment phase, covering all processes required to discharge, dismantle, and shred the batteries into fractions suitable for downstream recovery. Processes related to collection, transportation, and final material recovery (recycling) are excluded, as they fall outside the scope of this study. The functional unit for the analysis is defined as one metric tonne of spent LIBs processed through the pre-treatment phase, ensuring comparability with similar studies and practical relevance to the recycling industry. The scope of this study and system boundaries within the whole LIB life cycle are highlighted in red and shown in Fig. 1 (c). The processes from the Ecoinvent database used for LCA modeling are presented in Table 3. Each process was carefully selected to align with the defined system boundaries and to ensure consistency in representing the material flows, energy inputs, and emissions specific to the studied facility. These processes were carefully selected to provide a foundation for the analysis and were subsequently transformed and modified based on the available inventory data to better align with the specific conditions of the study, ensuring that the modeling reflects the most accurate and relevant environmental impacts.

Environmental impact categories (EICs) can vary depending on the context of the assessment and the specific goals of the study (EF3.1 Impact Categories | Earthster Knowledge Base, 2024). Commonly recognized EICs include climate change, resource depletion, water pollution, air pollution, ecotoxicity, land use, and waste generation. For this study, three main EICs were identified: climate change, eutrophication, and resource use (specifically minerals and metals). These categories were selected based on their relevance to the environmental performance of LIB pre-treatment processes and their alignment with key regulatory and industrial priorities. To enhance the reliability of results, normalization and sensitivity analyses were conducted, ensuring that the selected categories adequately capture the critical environmental impacts of the studied system. Chosen categories were based on normalized results (Klöpffer and Grahl, 2014).

The Intergovernmental Panel on Climate Change (IPCC) characterization model is used to evaluate the impacts of climate change. This model provides characterization factors quantified as the Global Warming Potential over a 100-year time frame (GWP100), expressed in kilograms of carbon dioxide equivalent (kg CO2 eq.) per kilogram of emission. The indicator operates on a global scale. The GWP data reported by the IPCC are specifically related to emissions to the air. To align these values with the classifications used in the ILCD (International Life Cycle Data system) and EF (Environmental Footprint) frameworks, they have been distributed across various emission compartments, including emissions to the lower stratosphere and upper troposphere, emissions to non-urban air or from high stacks, and emissions to urban air close to the ground (Glasson and Therivel; Klöpffer and Grahl, 2014; Myhre G. et al., 2013).

Eutrophication, freshwater EIC measures the degree to which freshwater ecosystems become overly enriched with nutrients, stemming from emissions of compounds containing phosphorus. This process encompasses all effects linked to the excessive presence of macronutrients in the environment, originating from nutrient emissions to air, water, and soil. Additionally, it represents the expression of the degree to which the emitted nutrients reach the freshwater end compartment (phosphorus is considered a limiting factor in freshwater) (Struijs et al., 2009).

Resource use, minerals and metals EIC focuses on the consumption of natural non-fossil resources, specifically minerals and metals. The indicator is associated with the extraction of minerals and fossil fuels as inputs into various systems. The Abiotic Depletion Factor (ADF) calculates the impact of each mineral extraction, expressed in kilograms of antimony equivalents (Sb eq.), considering reserve concentrations and the rate of depletion. This indicator's applicability is global, encompassing the entire planet (Glasson & Therivel, n.d.; Klöpffer and Grahl, 2014; Van Oers et al., 2016).

3. Results and discussion

3.1. Scenario 2022; 2030 comparison

The comprehensive assessment of this study involved calculating the annual environmental impacts for selected operational scenarios and comparing the impacts based on the energy mixes for 2022 and 2030. A summary of these impacts is provided in the SM. The results highlight that the recycling pre-treatment of spent LIBs has environmental benefits (negative sign of the environmental impacts) due to assumption that the recycled materials replace production of virgin materials, which environmental impacts are deducted. This approach is commonly applied in LCA under the concept of avoided products (the production of virgin materials is avoided due to recycled materials). Fig. 2 illustrates the comparison of case study scenarios modelled with different energy mixes for 2022 and 2030.

The comparison indicates that recycling EV batteries results in higher environmental benefits for eutrophication, freshwater use, and resource use (including minerals and metals) compared to CECs. However, CECs offer higher environmental benefits for climate change than

Table 3
The inventory of Ecoinvent 3.9 processes used for the life cycle analysis modelling.

Ecovinvent 3.9 processes
Secondary materials (their production is considered as avoided product by materials recycled during pre-treatment process)
Polypropylene, granulate {RER} | polypropylene production, granulate | Cut-off, U
Polyethylene, high density, granulate {RER} | polyethylene production, high density, granulate | Cut-off, U
Pig iron {RER} | pig iron production | Cut-off, U
Aluminium, primary, ingot {RoW} | aluminium production, primary, ingot | Cut-off, U
Copper, cathode {GLO}| market for copper, cathode | Cut-off, U
Synthetic graphite, battery grade {RoW} | synthetic graphite production, battery grade | Cut-off, U
Waste products
Wastewater, average {Europe without Switzerland} | market for wastewater, average | Cut-off, U
Sewage sludge, 75% water, WWT, WW, average {Europe without Switzerland} | treatment of sewage sludge, 75% water, WWT, WW, average, sanitary landfill | Cut-off, U
Waste plastic plaster, for final disposal {RoW} | treatment of waste plastic plaster, inert material landfill | Cut-off, U
Other (machines, energy)
Industrial machine, heavy, unspecified {RER}| market for industrial machine, heavy, unspecified | Cut-off, U
Salt {GLO}| market for salt | Cut-off, U
Tap water {Europe without Switzerland} | tap water production, conventional treatment | Cut-off, U
Conveyor belt {RER} | conveyor belt production | Cut-off, U
Electricity, low voltage {CZ} | electricity voltage transformation from medium to low voltage | Cut-off, U
Auxiliary heating unit, electric, 5 kW {GLO} | market for auxiliary heating unit, electric, 5 kW | Cut-off, U

Note: RER – "Rest of Europe"; RoW – "Rest of the world"; GLO – "Global"; WWT – "Wastewater treatment"; WW – "Wastewater"; CZ – "Czech Republic".

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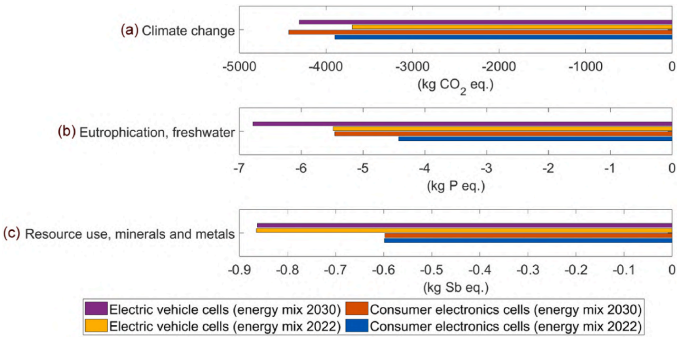

Fig. 2. Comparison of scenarios regarding energy mix compositions in 2022 and 2030, illustrating selected environmental impact categories: (a) climate change, (b) eutrophication, freshwater, and (c) resource use, minerals and metals. Results are presented per functional unit of one metric tonne of end-of-life lithium-ion batteries.

EV batteries. Specifically, this is attributed to the lower energy intensity of processing CEC batteries and the differences in avoided products derived from their material composition. Using the 2022 energy mix, CECs demonstrate environmental benefits of 80% in eutrophication, freshwater, and 69% in resource use, minerals, and metals relative to EV batteries. In contrast, EV batteries have 5% lower benefits for climate change compared to CECs.

The differences in resource use, minerals and metals EIC between CECs and EV cells arise mainly from variations in the internal material ratios, such as cathode active materials. For example, EV batteries often rely on cobalt- and nickel-rich cathodes (e.g., NMC), which significantly contribute to resource depletion metrics, while CECs commonly use materials with lower criticality, such as manganese-based cathodes (e.g., LMO). These differences are key contributors to the environmental impacts observed in LIB recycling. Proper processing of CECs is crucial for environmental sustainability, highlighting the importance of addressing CEC waste in recycling pre-treatment efforts.

Additionally, despite variations in energy mixes, the production technology of avoided products (such as Al and Cu) remains constant across the scenarios. This assumption reflects the current state of metallurgical recovery technologies and their efficiency, which are assumed not to change significantly in the short term. The comparison reveals that the energy mix significantly affects eutrophication and freshwater EICs (>20%), has a moderate impact on climate change EIC (<17%), and a minimal effect on resource use, minerals, and metals EIC (<1%). These results underline the role of energy transition in shaping the environmental benefits of recycling technologies, particularly for impact categories sensitive to energy sources.

Overall, the 2030 scenario shows a substantial improvement in environmental benefits for the first two categories, mainly due to the reduced reliance on coal-fired power plants, which constituted nearly half of the Czech Republic's energy mix in 2022 but is expected to drop to less than a third by 2030. This improvement underscores the importance of aligning recycling strategies with cleaner energy systems to maximize environmental performance. In contrast, the benefits related to resource use, minerals, and metals EIC show a decline, with only a marginal change of 0.20% observed in comparison to treated battery cell technologies.

3.1.1. A detailed breakdown within Scenario 2022

To offer a comprehensive understanding of the influence and characteristics of the output product, a detailed breakdown was created for Scenario 2022, as shown in Fig. 3. This breakdown includes the processing of output materials across various impact categories: recycling line, waste binders, electrolyte (including electricity used in plant operations), wastewater treatment, and secondary plastics, secondary iron and steel, secondary aluminium, secondary copper, and secondary graphite. These categories were selected based on their relevance to the key environmental impacts associated with the recycling process, as well as their contribution to the material recovery system. The impacts of these categories and their total contributions to the selected EICs were analysed and determined.

The analysed EICs reflect the environmental impact of further processing of materials within the output flows. For disposal-related categories, such as recycling line, waste binders, electrolyte, and wastewater treatment, environmental impacts were assessed across all categories. This is largely a result of the Czech energy mix, where over 40% derived from fossil fuels, powering the recycling process. This underscores the critical role of energy sources in shaping the environmental outcomes of recycling activities, highlighting the importance of integrating cleaner energy alternatives into future recycling technologies. In contrast, the reuse of materials, represented by the "secondary" categories, results in negative values for environmental impacts, thus reducing overall impacts and providing environmental benefits. Such benefits assume that secondary materials, once recovered, displace the need for primary extraction and production processes, leading to a reduction in environmental impacts across multiple categories. Wastewater processing was not considered in the context of EV battery unit treatment in this study, leading to zero impact in all selected EICs. However, the exclusion of wastewater treatment should be acknowledged as a limitation in the assessment of water-related impacts. Details of the pre-treatment steps are illustrated in Fig. 1(a).

The categories "secondary copper" and "secondary aluminium" significantly impact the analysis because their use in place of primary production leads to the deduction of potential environmental impacts, thanks to the assumed 100% recovery rate for these metals. While aluminium's impact on the resource use, minerals, and metals EIC is minimal due to its abundance, its high-energy production process has a major effect on climate change. This highlights the trade-off between the resource availability of a material and the energy required for its recovery. Conversely, copper has a more substantial impact on metal depletion. Given its limited natural reserves and high demand in industries beyond batteries, copper recovery plays a key role in reducing metal depletion. The environmental benefits of recovering secondary plastic parts, steel, and iron from consumer electronics cells and electric vehicle cells are comparable, as both types of battery cells contain similar proportions of these materials. However, the benefits of recovering these materials are contingent on the processing efficiency and the quality of the recycling technologies employed.

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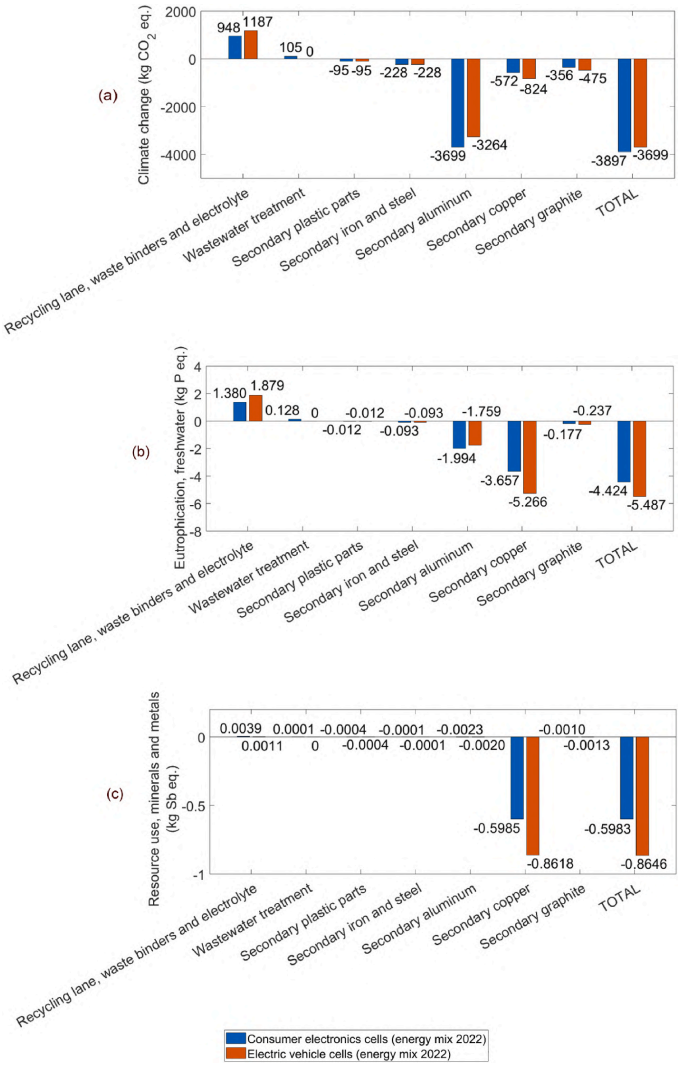

Fig. 3. A detailed breakdown of environmental impact categories within Scenario 2022, a) climate change, b) eutrophication, freshwater, and c) resource use, minerals and metals.

3.2. Sensitivity analysis

Two sensitivity analyses are provided in this study. SA1 examines the impact of technology efficiency, while SA2 focuses on the effect of secondary material usage on selected EICs. The results are organized by battery cell type, with EV cells shown in Figure (a) and CECs in Figure (b). Variants are listed alphabetically and arranged from least to most efficient in terms of technology or secondary material usage. The variant demonstrating the greatest environmental benefit, marked by the most negative EIC value, is considered the baseline at 100%. All other variants are evaluated relative to this baseline to determine their impact and changes. Additional details are available in Table 2.

3.2.1. Technology efficiency impact

The impacts of all monitored EICs for both battery technologies are contingent upon the effectiveness of the technology employed. As technology efficiency improves, the potential for mitigating environmental impacts increases, highlighting the crucial role of process optimization. All analysed scenarios lead to negative impacts; nevertheless, as the efficiency of the technology decreases, so does the ability to

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reduce or mitigate environmental impacts. The results of SA1 are illustrated in detail in Fig. 4.

Analysing both battery types reveals differences in EIC changes as technology efficiencies vary. For both EV batteries and CECs, differences between variants C and D were negligible (up to 2%) due to a 20% increase in efficiency for the recycling of other metals from 80% to 100%. The most significant changes occurred when the recycling efficiency for Al and Cu foils increased to 100%. Significant improvements were noted when the recycling efficiency of high-impact materials such as Al and Cu foils was optimized. In comparing variant D to the "energy mix 2022″ scenario, substantial improvements were observed, with reductions in EICs ranging from 20% to 25% for climate change, eutrophication, freshwater, and resource use, minerals and metals. For CECs, the reductions were similarly substantial.

The difference between variants A and B, involving the processing of plastics and binders, was minimal across both battery types, particularly for CECs. The difference in resource use, minerals, and metals EIC is negligible, as the impact of plastics and binders on this category is limited. Even with a 100% increase in processing efficiency for these materials, the change in overall EICs remains modest, with the greatest reductions observed in climate change impacts. Climate change impacts showing a range of approximately 3–8% for EVs and negligible effects (up to 2%) on other categories for CECs. In both pre-treatment recycling pathways, notable differences were found between variants B and C when graphite processing efficiency increased to 100%. This led to 6–8% changes in climate change and negligible impacts (up to 2%) for eutrophication, freshwater, and resource use, minerals and metals in EV cells and CECs recycling route.

The SA1 results emphasize that nearly all materials are either recycled or landfilled, demonstrating the benefits of effective secondary treatment in LIB recycling for EICs, especially climate change, eutrophication, freshwater and resource use, minerals and metals. The efficiency of Al and Cu foil recovery plays a pivotal role in achieving the largest environmental benefits. At the same time, changes in the recycling efficiency of plastics and binders have had a negligible effect on all monitored EICs.

3.1.2. Secondary material usage impact

The SA2 results highlight the critical role of secondary material used in recycling pre-treatment, significantly affecting environmental impacts, as shown in Fig. 5. The most negligible differences between variants Y and the "energy mix 2022″ scenario were observed for both EV cells and CECs, comparing 100% secondary use with complete recycling. For EV cells, the differences were negligible (up to 2%) for climate

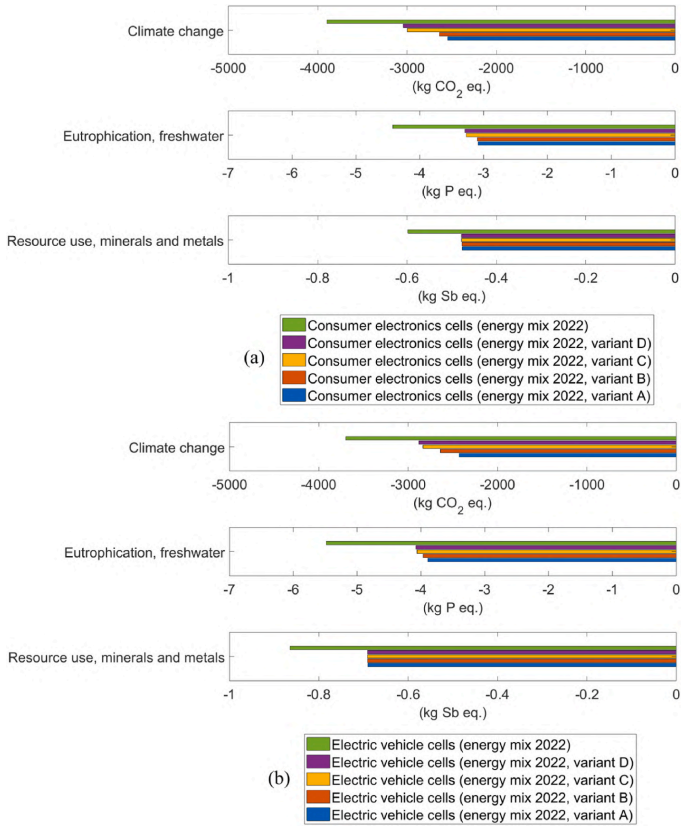

Fig. 4. Sensitivity analysis 1 results describing the effect of efficiency of implemented technology for (a) consumer electronics cells and (b) electric vehicle cells. Presented results reflect the standardized comparison with the assistance of a functional unit (one metric tonne of end-of-life lithium-ion batteries).

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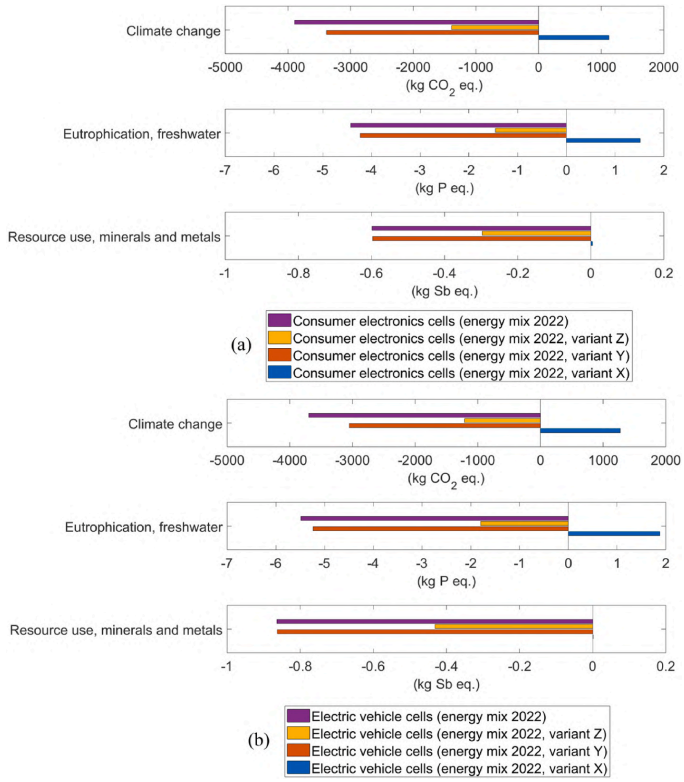

Fig. 5. Sensitivity analysis 2 results address the effect of the secondary material impact usage for (a) consumer electronics cells and (b) electric vehicle cells. Presented results reflect the standardized comparison with the assistance of a functional unit (one metric tonne of end-of-life lithium-ion batteries).

change, and eutrophication, freshwater impacts, while the resource use, minerals and metals impact was around 0.2%. For CECs, the differences were similarly minimal, with negligible changes for climate change and eutrophication, freshwater and approximately 0.2% for resource use, minerals and metals.

Variant Z, involving 50% material reuse, shows moderate reductions in environmental impacts compared to complete processing with "energy mix 2022." For EV cells, reductions are about 67% for both climate change, and eutrophication, freshwater, and around 50% for resource use, minerals, and metals. For CECs, the reductions are similarly significant, with approximately 64% for climate change and around 50% for resource use, minerals and metals.

In contrast, variant X, with no secondary processing, worsens impacts across all categories, emphasizing the importance of secondary material use. Without secondary processing, the environmental benefits of recycling are severely diminished. For both battery types, the rise in the resource use impact category is minimal; however, the increase in climate change is notable. For EVs, the impact rises from −3700 kg CO_2 eq. to 1300 kg CO_2 eq., and for CECs, it increases from −3900 kg CO_2 eq. to 1100 kg CO_2 eq. Similarly, eutrophication and freshwater impacts increase: for EVs, from −5.5 kg CO_2 eq. to 1.9 kg CO_2 eq., and for CECs, from −4.4 kg CO_2 eq. to 1.5 kg CO_2 eq. These findings underscore the essential role of secondary metal use in achieving significant environmental benefits. Without it, the overall advantages of recycling are severely diminished.

3.2.3. *Sensitivity analysis results*

When comparing the results from both the SA1 and SA2 analyses, it becomes clear that secondary material use is a crucial factor in the recycling and waste treatment of LIBs, regardless of whether the batteries are EV cells or CECs. Even with low-efficiency technology, characterized by poor material separation and reduced yield, the 100% reuse of recovered materials still led to environmentally favourable outcomes, reducing the impact of individual EICs by 17%–64%. This highlights the importance of optimizing material recovery processes and demonstrates that effective secondary material utilization is a key strategy for improving environmental performance, even under suboptimal technological conditions.

Conversely, even when using high-quality technology capable of recovering up to 100% of battery materials, neglecting the secondary use of these materials can still result in significant environmental impacts. For example, the "energy efficiency 2022″ variant demonstrates that failing to reuse recovered materials can increase environmental impacts by as much as 35%. This finding underscores the importance of implementing comprehensive material recovery strategies, where not only the technological efficiency but also the post-recovery utilization phase plays a pivotal role in determining the overall environmental benefits.

The most significant benefits from secondary metal recovery are observed in the climate change impact category, primarily due to the high energy requirements of primary metal production. However, consistent achievement of high recovery rates is contingent upon advancements in both technology and economic feasibility, as not all facilities are equipped to employ the most efficient methods. Technological improvements in material separation, coupled with economic incentives for secondary material use, are essential for further enhancing the environmental sustainability of LIB recycling.

4. Conclusion

This study presents a comprehensive gate-to-gate LCA of LIB recycling pre-treatment at a small-scale facility in the Czech Republic, targeting EVs and CECs. The results are highly relevant for policymakers, recycling facility operators, and researchers focused on sustainable energy and material recovery. The findings highlight significant environmental benefits, particularly in reducing impacts on climate change, eutrophication, freshwater, resource use, minerals and metals. Notably, CEC recycling demonstrates greater effectiveness in climate change mitigation than EV battery recycling. This observation underscores the importance of developing tailored strategies for different battery types to optimize their recycling processes and maximize environmental benefits.

A key takeaway is the critical role of secondary material recovery, especially for metals like copper and aluminium, which substantially reduce resource use and emissions. Achieving high recovery efficiency is essential not only for material conservation but also for minimizing environmental impacts across multiple impact categories. To this end, recycling facilities must optimize technological processes to enhance material recovery and minimize environmental impacts.

To better contextualize the significance of the recycling pre-treatment, the environmental benefits were compared to the impacts associated with the production of batteries. In the climate change impact category, recycling results in a 35% reduction in environmental impact for CECs and 33% for EV batteries relative to their production impacts. In the eutrophication, freshwater category, recycling pre-treatment achieves a 45% reduction for CECs and 55% for EV batteries. Regarding resource use, minerals and metals, the environmental benefits of recycling are 38% for CECs and 55% for EV batteries. These findings highlight that the environmental advantages of recycling pre-treatment significantly outweigh the impacts of battery production. Consequently, LCA studies of technologies utilizing batteries that do not account for their recycling at the end of life may greatly exaggerate the environmental impacts of these technologies.

The analysis further highlights the critical role of a cleaner energy mix in reducing environmental impacts. As the energy grid transitions to lower-carbon sources, reductions in impacts, particularly eutrophication and freshwater use, are expected. This indicates that policy-driven transitions in the energy mix will further optimize LIB recycling processes, especially in terms of their environmental benefits. However, regardless of the energy scenario, efficient secondary material recovery remains indispensable to achieving optimal environmental outcomes.

While this study emphasizes the advantages of current pre-treatment methods, uncertainties regarding battery composition and the variable performance of recycling technologies remain. Expanding the data set to include a broader range of LIB chemistries and recycling methods would provide a more robust understanding of environmental impacts. A thorough assessment of specific recycling technologies, including pyrometallurgical, hydrometallurgical, and direct physical separation methods, would provide valuable insights into process efficiencies and environmental trade-offs. Furthermore, this study fills a notable gap in the literature by focusing on the pre-treatment phase, a critical yet often overlooked component of the LIB recycling process. The insights gained from this research establish a foundation for further comparative studies, providing an essential step towards more sustainable recycling practices.

In conclusion, this study offers critical insights into optimizing LIB recycling processes, establishing a framework for enhancing recycling technologies and maximizing material recovery. Future research should aim to address existing data gaps and broaden LCA coverage to include a diverse array of battery chemistries and recycling pathways. Expanding the scope of future research will not only deepen our understanding of the pre-treatment process but also help optimize recycling strategies for a broader range of battery technologies, contributing to more sustainable energy transitions.

CRediT authorship contribution statement

Anna Pražanová: Writing – original draft, Resources, Project administration, Methodology, Conceptualization. **Michael Fridrich:** Visualization, Investigation, Formal analysis, Data curation. **Jan Weinzettel:** Writing – review & editing, Supervision. **Vaclav Knap:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Anna Prazanova reports financial support was provided by Czech Technical University in Prague. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cesys.2025.100263.

Data availability

Data will be made available on request.

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Environmental and Economic Assessment of Battery Energy Storage System Disposal and Recycling Treatment in the Czech Republic

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Keywords: lithium-ion battery, battery energy storage system, end-of-life, recycling pre-treatment, environmental perspective, economic evaluation

Abstract
As global energy systems transition towards decarbonization and increased reliance on renewable resources, the need for effective solutions to manage the lifecycle of energy storage systems becomes increasingly critical. Battery energy storage systems (BESSs) have emerged as key players in ensuring grid stability and supporting the integration of intermittent renewable energy sources. Yet, their widespread adoption faces significant environmental and economic challenges, particularly concerning the end-of-life (EOL) management of lithium-ion batteries (LIBs). This work presents an environmental and economic evaluation of a 2.8 MWh/2.5 MW BESS case study, specifically focusing on EOL treatment strategies. The study employs a life cycle assessment (LCA) to examine recycling pre-treatment processes for the battery, cooling, and extinguishing systems, inverters, and reuse of BESS containers and substations. Methodologically, the LCA assesses the environmental benefits of recovering secondary materials, particularly aluminium and copper, revealing that recycling pre-treatment processes contribute to one-third of the overall environmental gains. The economic analysis projects

potential profits of around €69,000 per unit, highlighting the significant financial incentives for local sourcing of critical raw materials. Beyond the financial and environmental implications, the study underscores the importance of sustainable recycling practices and presents opportunities for enhancing self-sufficiency in critical materials. These findings emphasize the relevance of efficient EOL management and provide insights for improving environmental and economic strategies in BESS management, offering valuable guidance for future research, policy development, and industry practices.

1. Introduction

Energy systems are undergoing a global transformation driven by technological advancements, decarbonization efforts, the deployment of smart grids, and the increasing integration of renewable energy resources. Historically reliant on fossil fuels, power generation has significantly contributed to greenhouse gas (GHG) emissions and global warming. To address this, efforts focus on reducing emissions and expanding sustainable energy sources (Intergovernmental Panel on Climate Change (IPCC), 2023; IRENA, 2023). Decentralized power generation and autonomous microgrids, however, pose challenges for grid infrastructure (Nebey, 2024). Battery energy storage systems (BESSs) play a critical role by storing and releasing energy to ensure grid stability and reliability. They are used for load levelling, grid frequency regulation, and backup power during outages. Additionally, BESSs are invaluable for integrating intermittent renewable sources like solar and wind, ensuring consistent supply (IRENA, 2017; Zakeri & Syri, 2015). They also reduce the need for costly infrastructure upgrades, such as peaking power plants and enhanced transmission networks (Divya & Østergaard, 2009; Hannan et al., 2021). Key challenges include high costs, limited lifespan, and environmental impacts, with battery degradation posing scalability concerns for large-scale storage. Despite these issues, BESSs are increasingly adopted for renewable energy integration and grid support. Advancing technology and declining costs make them vital for a sustainable energy future (Hannan et al., 2021; Hengky K. Salim et al., 2019). As adoption grows, addressing end-of-life (EOL) treatment becomes crucial (Toro et al., 2023; Zhang, 2023).

Waste BESSs treatment is crucial for sustainable management, as it facilitates resource recovery, reduces environmental impacts, and supports the transition to a circular economy (X. Liu et al., 2023; Ndiribe, 2023; Pinto et al., 2024; Sulemanu, 2023). These systems typically operate for 10 to 20 years, similar to the maximum 10-year lifespan of EV batteries (Ohrelius et al., 2023). Given their similar technology, there is interest in repurposing EOL modules from EVs for BESS installations, extending battery life and enhancing resource efficiency. Efficient recycling and recovery processes for BESS components allow for extracting valuable materials and reducing the environmental footprint (Larcher & Tarascon, 2015; Sulemanu, 2023). Currently, BESSs used in both small-scale and large industrial systems, are mostly based on the lithium-ion battery (LIB) technology (Hannan et al., 2021; Ohrelius et al., 2023). The high proportion of rare materials like cobalt, nickel, manganese, and lithium in these batteries underscores the importance of efficient EOL management to maximize resource utilization and sustainability (Conzen et al., 2023).

These battery cells consist of active layers made from materials such as lithium cobalt oxide ($LiCoO_2$), lithium iron phosphate ($LiFePO_4$), nickel cobalt aluminium (NCA), or nickel manganese cobalt oxide (NMC) deposited on aluminium foils, which serve as the current collectors for the positive electrodes. The negative electrodes typically comprise a graphite layer deposited on copper current collectors. Additionally, the electrolyte typically contains lithium hexafluorophosphate ($LiPF_6$) salt dissolved in organic solvents, which enables the movement of lithium ions through polymer separators between the electrodes during charging and discharging cycles (Pražanová et al., 2022a; Thompson et al., 2020; Zhu et al., 2021).

Individual battery cells are combined into modules, which act as the fundamental BESSs building blocks. These modules arrange cells in series and parallel to meet the desired voltage, capacity, and performance. In BESS installations, modules are arranged in arrays, interconnected with power electronics and control systems for overall operation (Chang et al., 2018; Ovaskainen et al., 2023). This modular design facilitates scalability and flexibility, allowing for the creation of BESSs tailored to various energy storage needs, whether for small-scale residential applications or large-scale industrial deployments supporting grid stability and renewable energy integration (Conzen et al., 2023; IRENA, 2023; Pinto et al., 2024). However, despite the advancements in materials over the past three decades, the fundamental design of these battery cells has largely remained unchanged. This poses significant challenges for recycling, as the current design does not prioritize recyclability, thereby limiting the long-term sustainability of battery systems. Addressing these challenges is essential for ensuring that future battery technologies are both effective in energy storage and environmentally responsible.

In addition to battery modules, BESSs include essential components like power converters for managing flow and grid interfacing, thermal management systems (TMS) for maintaining optimal battery temperatures, fire extinguishing systems for emergencies, and energy management systems (EMS) for monitoring and controlling charging, discharging, and overall performance. The enclosure or container provides structural support and protection. Material-wise, inverters use steel and copper, along with aluminium electrolytic or ceramic capacitors, copper-based inductors, and copper- or gold-traced (PCBs). The TMS includes heat sinks, heat pipes, ceramic or polymer insulation, and nitrogen-based fire suppression. The EMS integrates microcontrollers, sensors, and semiconductors. Containers are typically made from steel, aluminium, or composites, and some installations include substations with similar materials (Chang et al., 2018; Hossain et al., 2019; Jasper et al., 2022; H. K. Salim et al., 2020).

Battery systems present unique challenges in waste processing compared to other components typically handled through metal recovery or repair. Processing batteries involves disassembling modules from racks, deep discharging to prevent ignition, and crushing to separate materials like current collectors, active electrode materials, and packaging. Thermal treatment removes organic components such as electrolytes and binders, producing black mass—a valuable mix of metals central to recycling. Black mass is processed via pyrometallurgy (high-temperature recovery) or hydrometallurgy (chemical solutions), recovering elements like lithium, cobalt, nickel, and manganese for reuse in battery manufacturing. Efficient pre-treatment is critical for consistently producing high-quality black mass (Chang et al., 2018; Hossain et al., 2019; Li Li et al., 2018; S. Liu et al., 2022; Anna Pražanová et al., 2022a; Toro et al., 2023).

The EOL treatment of LIBs presents significant challenges. Recycling and pre-treatment methods are energy-intensive and financially demanding (Li et al., 2018). Additionally, not all technologies achieve full material recovery and may generate significant waste or hazardous by-products (Hayes & McCullough, 2018). The diversity of recycling operations complicates the process, as each step, from disassembly to material recovery, varies in efficiency and environmental impacts. Generalizing about LIB recycling technologies is therefore not feasible (Chen et al., 2019; Hayes & McCullough, 2018; Natarajan & Aravindan, 2018; Sommerville et al., 2021). Each process must be individually evaluated, considering its specific steps and methods. This work addresses these gaps by assessing the environmental and economic viability of EOL treatment processes with a case study emphasizing the importance of process-specific evaluations.

The environmental impacts of EOL treatment for LIB-based BESSs are significant, particularly in recycling pre-treatment. These processes simplify recycling and reduce environmental footprints but pose challenges due to their energy-intensive nature (Pražanová et al., 2025). Romare & Dahllöf (2017) highlight that energy consumption during disassembly

and discharge significantly affects the environmental performance of battery recycling. Additionally, crushing and material separation generate dust and hazardous emissions, further increasing the environmental burden. Bobba et al. (2018) note that pre-treatment processes, involving high-energy machinery and chemical treatments, contribute substantially to GHG emissions and resource consumption. Despite these challenges, pre-treatment effectively recovers valuable materials, reducing reliance on virgin resources. This study evaluates trade-offs between environmental impacts and material recovery efficiency, offering insights into the environmental implications of these processes.

The economic aspects of recycling are complex, primarily due to high initial costs for disassembly, deep discharge, and processing, as noted by Harper et al. (2019). Energy-intensive pre-treatment steps, such as crushing and separation, further increase operational expenses. Fluctuating market prices for recovered materials like lithium, cobalt, and nickel impact profitability (Larcher & Tarascon, 2015). Wang et al. (2014) emphasize that efficient processes yield high-purity materials for new batteries, reducing raw material mining and stabilizing supply chains. Advancements in technology, along with economies of scale, can help reduce costs (Gaines, 2018). Moreover, developing recycling infrastructure can generate new jobs and spur economic growth, as Hayes & McCullough (2018) suggest.

This study evaluates the environmental and economic impacts of EOL treatment for a 2.8 MWh/2.5 MW LIB-based BESS. It investigates battery recycling pre-treatment, the treatment of cooling and extinguishing systems, inverters, and the reuse of containers and substations in new installations. Using a gate-to-gate life cycle assessment (LCA), the study quantifies the significant environmental benefits of reusing secondary materials like aluminium, copper, and steel, offering a more comprehensive analysis than previous research. Economically, BESS recycling demonstrates clear benefits despite the added costs of supplementary technologies. The study also highlights the economic advantages of local raw material production, including reduced import dependency, improved supply chain security, and lower transportation costs and emissions. The findings underscore the dual benefits of BESS recycling: environmental improvements through material reuse and economic gains. Additionally, transitioning to renewable energy sources for recycling could amplify these benefits, supporting broader sustainability goals and advancing more sustainable recycling practices.

2. Materials and methods

2.1 Goal, scope, and assumptions

This study evaluates the environmental and economic impacts of a BESS EOL treatment case in the Czech Republic. The system, designed for frequency regulation services, has a 2.8 MWh capacity and a peak power of 2.5 MW, meeting typical regional standards for medium-sized energy storage applications (IBG Česko s.r.o., 2019; Prakash et al., 2022; Siemens s.r.o., 2019).

The BESS comprises six components: battery system, thermal management, extinguishing system, inverters, container, and distribution board. This study focuses on recycling or material recovery from the battery, thermal management, extinguishing systems, and inverters. The container and substation, with longer lifespans and reuse potential, fall outside the scope, as they pertain to second-life applications (**Figure 1a**).

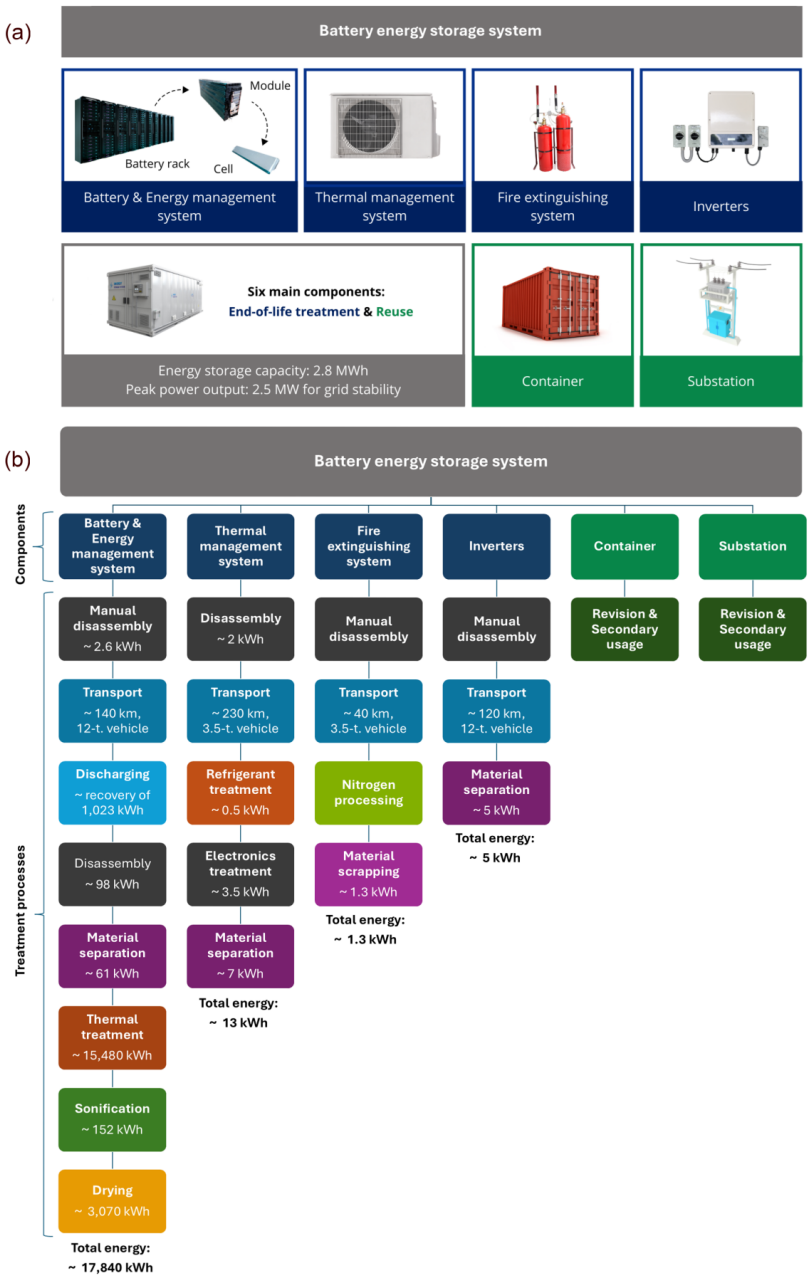

Figure 1. (a) Illustration of the battery energy storage system (BESS) components categorized by their respective treatments considered in the study. (b) Process characterization of the treatment steps for individual components, detailing material and energy flows.

The BESS unit, represented as a single container, consists of 409 LIB modules, which together house a total of 9,816 pouch cells based on NMC chemistry (IBG Česko s.r.o., 2019; A. Pražanová et al., 2022). Designed for waste treatment, the entire battery system weighs 13,088 kg and comprises these 409 modules, each with a weight of 32 kg. Within each module, there are 24 battery cells, weighing 1.1 kg each, along with a cover that adds 4.7 kg and binding materials totalling 0.91 kg. Each battery cell's composition includes approximately 44% positive electrode active materials (based on NMC 6.5:2:1.5), 32% graphite, 6% copper, 7.5% aluminium, 4.5% aluminium-polymer casing, and 6% organic and plastic parts, which encompass the separator, binders, and electrolyte (Anna Pražanová et al., 2023). In this study, "black mass" refers specifically to a product composed solely of positive electrode active materials. Generally, black mass can be understood as a mixture of valuable battery materials (from both electrodes, along with remnants of current collectors or casing materials). Given the nature of the applied method and the ability to directly recover selected materials, the black mass in this work is exclusively composed of NMC-based spinel

The electrical infrastructure includes four transformer-less hybrid inverters rated at 630 kW (Megarevo, 2024; Selvamathi et al., 2018) and one low-voltage distribution board (HIVE Connective Intelligence, 2023). The inverters collectively weigh 3,800 kg and comprise 1,140 kg of iron, 1,520 kg of aluminium, and 1,140 kg of electronic components, making them crucial for efficient energy conversion. Two cooling units with a heat output capacity of 12.5 kW are used to regulate the system's temperature (HIVE Connective Intelligence, 2023). The cooling system weighs a total of 490 kg and consists of 181.3 kg of iron (to be processed as pig iron), 63.7 kg of aluminium, 73.5 kg of copper, 28 kg of brass, 122.5 kg of plastics, 14 kg of electronic elements, and 7 kg of refrigerants. Additionally, fire safety measures, including an extinguishing system, are in place, based on based on four steel pressure cylinders, each weighing 70.2 kg, filled with 23.4 kg of nitrogen gas to ensure proper gas distribution for fire suppression (Global, 2017). All these components are housed in a metal shipping container with suitable accessories to support the system's operation and safety (HIVE Connective Intelligence, 2023). The weights of the processed materials for the individual components of the entire BESS are shown in Table 1. Considering an optimistic scenario and the exceptionally high efficiency of the technology, the input materials also represent the values of the intermediate or resulting materials obtained from the assessed processes.

The dismantling of the BESS begins with the removal of the battery system, where individual modules are manually disassembled from their racks and subsequently transported to the recycling facility. Upon arrival, the modules undergo a discharge process to recover any residual energy, after which the duralumin covers are milled off. The pouch cells, now separated from the cover residue, are subjected to robotic disassembly. During this stage, the electrode sets are sorted into cathodes and anodes, enabling further processing of these materials. The sorted components are then sheared and crushed before being fired in a furnace to enhance material recovery. In this work, it is assumed that all lithium is lost during this process. Following the firing process, current collectors are cleaned using ultrasonic baths, while filtration methods are employed to separate cleaned metal foils from the active materials. Any residual water is removed from the solid residues via centrifugation, after which the materials are dried and prepared for further treatment. Material handling throughout this process is efficiently managed by an electric forklift.

	Battery system		Cooling system		Extinguishing system		Inverters	
	Material	Mass (kg)	Material	Mass (kg)	Material	Mass (kg)	Material	Mass (kg)
Module	Duralumin cover	1,922	Iron	181.3	Steel	280.8	Iron	1,140
	Organics (binders, plastics)	372	Aluminium	63.7	Nitrogen	93.64	Aluminium	1,520
	Copper	661	Copper	73.5			Electronics	1,140
	Aluminium	812	Brass	28				
Cell	Positive electrode active materials	4,721	Electronics	14				
	Graphite	3,465	Refrigerator	7				
	Aluminium-polymer casing	491	Plastics	122.5				
	Organics (electrolyte, separator, binders, plastics)	643						
Total mass (kg)		13,088		490		374		3,800

Table 1. The summary of material composition of the evaluated components in the battery energy storage system (BESS) including the battery system, cooling system, fire extinguishing system, and inverters. For the positive electrode active materials based on NMC 6.5:2:1.5, the distribution of elements is as follows: 65% nickel, 15% manganese, and 20% cobalt. The total amounts of these materials within the entire battery system are 3,069 kg of nickel, 944 kg of manganese, and 708 kg of cobalt. (HIVE Connective Intelligence, 2023; IBG Česko s.r.o., 2019; Megarevo, 2024; Anna Pražanová et al., 2023)

In addition to the battery system, cooling units are dismantled, with the fluorocarbon-based medium extracted before the manual disassembly of electronic parts. The remaining materials are crushed, with iron components magnetically extracted and non-ferrous metals sorted electrostatically. Fire extinguishing systems are similarly disassembled, and nitrogen and steel bottles are processed for reuse. Inverters follow the same process, allowing for the extraction of aluminium, pig iron, and electronic scraps. **Figure 1(b)** illustrates the treatment steps, and detailed information on consumption and equipment use is provided in the Supplementary materials (SMs).

In this study, the baseline scenario was defined with the assumption of maximum yield and processing efficiency for all treated components. This perspective posits that the processes are entirely lossless concerning materials designated for secondary use or further processing. Such an optimistic scenario serves as an idealized framework for evaluating the processes at their highest efficiency, thereby providing valuable insights into the lowest environmental impacts and the maximum potential profits that can be achieved through processing. To enhance the realism of the assessment, Overall Equipment Effectiveness (OEE) was incorporated into the analysis. This adjustment accounts for operational inefficiencies, including micro downtimes and intermediate processing steps, which often result in machines operating below their total capacity. By integrating OEE, the analysis not only more accurately reflects practical industrial conditions but also offers a comprehensive representation of real-world scenarios, facilitating a deeper understanding of the potential challenges and opportunities within the BESS treatment processes.

2.2 Environmental evaluation

This study conducts a partial gate-to-gate LCA (International Organization for Standardization, 2006) focusing on the EOL treatment of BESS, specifically examining recycling pre-treatment steps. It treats the used LIBs as burden-free, not including the environmental impacts from their previous lifecycle phases. The outputs obtained from this process are intended for further treatment. The assessment was performed using the 9.5 version of the SimaPro LCA modelling software, utilizing the Ecoinvent 3.9 database with a Cut-Off approach and the Environmental Footprint 3.1 (EF 3.1) Impact Assessment method. The scope and life-cycle boundaries of this study within the LIB life cycle are illustrated in **Figure 2**.

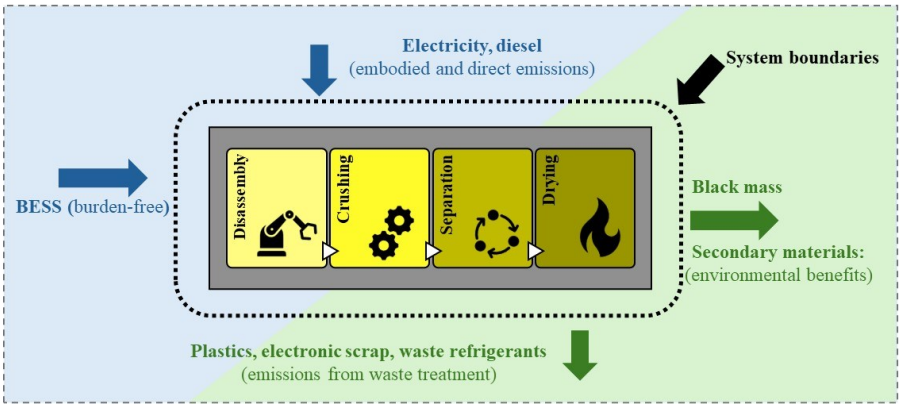

Figure 2. The scheme of system boundaries, which were considered for this partial LCA study. Study inputs (end-of-life batteries, energy) are shown in blue and outputs (recovered materials) in green.

This study evaluates the EOL treatment of four main BESS components: the LIB system, the cooling system with air-conditioning units, the fire extinguishing system, and the inverters. Each component underwent recycling, with the LIB system primarily producing black mass, a valuable mixture of materials. Significant secondary materials, including metals like aluminium, copper, and graphite, were recovered, highlighting the efficiency of the recycling process. The cooling system produced secondary aluminium, copper, iron, and brass. The fire extinguishing system provided secondary steel, while the inverter yielded secondary iron and aluminium. These materials will be reused, reducing the environmental impact by substituting primary materials. However, some materials were treated as waste in this process. Plastics from the batteries and cooling systems were incinerated. Electronic scrap was separated into steel and cables. The refrigerant from the cooling unit was treated separately, while nitrogen from the fire extinguishing system was released into the atmosphere.

The energy mix employed for operating the recycling machinery was based on the current Czech energy mix – fossil fuels (50.78 %), nuclear energy (42.82 %), renewables (6,4 %) (Czech electricity and gas market operator (OTE a.s.), 2024). Due to varying recycling methods and battery system compositions, this study quantifies environmental impacts per one recycled BESS container. This functional unit was selected to account for the complexity of the system and its components, including the battery system, inverters, thermal management, and extinguishing systems, which are designed to function as an integrated whole. While kWh-based functional units are commonly used for battery-specific analyses, they do not adequately capture the contribution of these infrastructure components, making them less suitable for

evaluating entire BESS systems. This approach enables clear comparisons between recycling pathways and allows for transparent analysis of each BESS component's contribution to the life cycle. More details of the evaluation using this functional unit in selected ICs are provided in the SMs of this work. A detailed examination of the Ecoinvent 3.9 processes used for modelling is provided in Table 2.

Ecovinvent 3.9 processes	Description
Avoided products (secondary materials)	
Aluminium, primary, ingot {RoW}| aluminium production, primary, ingot | Cut-off, U	secondary material
Copper, cathode {GLO}| market for copper, cathode | Cut-off, U	secondary material
Graphite, battery grade {RoW}| graphite production, battery grade | Cut-off, U	secondary material
Steel, chromium steel 18/8 {GLO}| market for steel, chromium steel 18/8 | Cut-off, U	secondary material
Pig iron {RER}| pig iron production | Cut-off, U	secondary material
Brass {RoW}| brass production | Cut-off, U	secondary material
Waste products (end-of-life)	
Waste plastic, mixture {RoW}| treatment of waste plastic, mixture, municipal incineration | Cut-off, U	waste plastic (batteries)
Waste plastic, consumer electronics {RoW}| treatment of waste plastic, consumer electronics, municipal incineration | Cut-off, U	waste plastic (AC)
Electronics scrap from control units {GLO}| market for electronics scrap from control units | Cut-off, U	waste electronics (AC, inverter)
Used refrigerant R134a {GLO}| treatment of used refrigerant R134a, reclamation | Cut-off, U	waste refrigerant
Other (energy, transport)	
Transport, freight, lorry 7.5-16 metric ton, EURO6 {RER}| market for transport, freight, lorry 7.5-16 metric ton, EURO6 | Cut-off, U	transport (batteries)
Industrial machine, heavy, unspecified {RER}| industrial machine production, heavy, unspecified | Cut-off, U	machines
Electricity, low voltage {CZ}| market for electricity, low voltage | Cut-off, U	Czech energy mix 2023
Diesel, low-sulfur {RER}| market group for diesel, low-sulfur | Cut-off, U	supply for crushing machine
Transport, freight, light commercial vehicle {Europe without Switzerland}| market for transport, freight, light commercial vehicle | Cut-off, U	transport (fire extinguisher, AC)
Transport, freight, lorry 3.5-7.5 metric ton, EURO6 {RER}| transport, freight, lorry 3.5-7.5 metric ton, EURO6 | Cut-off, U	transport (inverter)

Table 2. Ecoinvent 3.9 processes used for life cycle assessment modelling, used abbreviations: RoW = rest of the world, GLO = global, RER = rest of Europe.

The environmental impact categories (ICs) often vary based on the study's context and objectives but commonly include climate change, resource depletion, water pollution, air pollution, ecotoxicity, land use, and waste generation (Fazio et al., 2019). In this study, three primary ICs were identified: climate change, eutrophication, freshwater, and resource use, minerals and metals. These were chosen for their industry relevance and significant normalized results across all BESS parts.

The climate change impacts were evaluated using the Intergovernmental Panel on Climate Change (IPCC) characterization model, which calculates the Global Warming Potential over 100 years (GWP100) in kilograms of carbon dioxide equivalent per kilogram (kg CO_2 eq.) of emission. The GWP data, relevant to emissions to air, were distributed across various emission compartments to align with ILCD and EF frameworks. Eutrophication, freshwater IC measures nutrient enrichment in freshwater ecosystems, primarily from phosphorus emissions, leading to eutrophication. This impact is significant due to the role of phosphorus as a limiting factor in freshwater environments expressed in phosphorus equivalents (kg P eq.). Resource use, minerals, and metals IC focus on consuming natural non-fossil resources, evaluating the extraction impact using the Abiotic Depletion Factor (ADF), expressed in kilograms of antimony equivalents (Sb eq.). This considers reserve concentrations and depletion rates with global applicability (Fazio et al., 2019).

2.3 Economic analysis

When the BESS reaches the EOL phase, treatment can offer numerous advantages. The most significant benefit is its positive environmental impacts, as it promotes the circular economy and reduces the generation of non-recyclable waste. Furthermore, recycling enables using recycled materials instead of relying solely on extracting primary raw materials. While the environmental benefits of recycling are evident, it is crucial for the process also to be economically feasible. The cost and quality of recycled raw materials should be on par with or superior to those obtained from primary raw materials through mining (Beaudet et al., 2020; Pražanová et al., 2022a).

From an economic perspective, the recycling of BESS can be evaluated based on the potential returns derived from the value and quantity of recycled materials and the costs involved in the recycling and recovery processes. Revenue generation hinges on the market price of recycled raw materials, with certain metals such as aluminium, cobalt, copper, and nickel holding substantial value, particularly in LIBs. On the cost front, expenses can be categorized into four groups, encompassing transportation costs for conveying BESS and its components for recycling, machine costs indicative of the price of pre-owned machinery, labour costs, and energy consumption-related expenses (Liao et al., 2019; Pražanová et al., 2022b; Sun et al., 2020).

In the context of this research, revenues were analysed, considering the valuation of partial raw material outputs based on prevailing market prices of the corresponding commodities. The average market price of the given commodity from 1/1/2024 to 10/7/2024 was predominantly utilized to assess most materials. However, due to the unavailability of historical pricing data for certain raw materials (E. Zhu et al., 2024), current prices as of 10/7/2024 were employed for graphite and manganese. The valuation of steel and brass was aligned with the pricing of the relevant metal scrap, while the valuation of iron was tethered to the pricing of pig iron. Insight to used prices is provided in detail in SMs.

This study used a 100% efficient baseline scenario to define target conditions, which were recognized as idealized rather than realistic. Therefore, overall equipment effectiveness (OEE) was then factored into account for real-world conditions, acknowledging that machines will operate below full functionality due to micro downtimes and intermediate steps. This approach ensures that the analysis reflects realistic industrial scenarios.

For the analysis, costs were primarily determined by evaluating machine costs. This involved estimating the equipment's useful life (10 years), applying a 5% interest rate per year, and assuming 260 work shifts per year with 8-hour shifts. The purchase price and these assumptions were used to calculate the annual annuity payment, which was then divided by the number of shifts per year to determine the machine cost per shift, accounting for the time value of money. Additionally, the EOL treatment for the primary component of the BESS, specifically the batteries, was thoroughly assessed, considering the duration of the cycle and the number of

shifts required for its completion. Total machine costs were subsequently determined by multiplying the cost per shift by the number of shifts of the bottleneck machine, adjusted for its OEE. Furthermore, energy consumption for the entire BESS recycling process was estimated for the machinery and equipment used. The energy cost was derived by summing specific costs directly associated with electricity consumption, supplemented by fixed costs based on prevailing energy prices in the Czech Republic, budgeted for the relevant number of shifts.

The labour costs were calculated based on several factors, including the number of workers directly engaged in recycling, machine operation, and disassembling BESS and its sub-parts, as well as the appropriate number of shifts and the hourly wages for each employee. These cost calculations were based on the prevailing wages in blue-collar professions in the Czech Republic. Additionally, the expenses of transporting the BESS and its components were determined based on the distance travelled and the current per-kilometer transportation rates within the Czech Republic.

The total costs were calculated by aggregating machine, labour, transportation, and energy costs. This simplified approach attempts to account for the time value of money and the uninterrupted operation of machinery in the industrial process. Upon assessing the weight yield of the recycled materials and their market price, as well as the direct costs associated with their recycling, the potential earnings before interest and taxes (EBIT) were calculated. Then, the Czech Republic's VAT of 21% was applied to determine the tax burden, resulting in the net profit (EAT).

3. Results and discussion

In this study, the economic analysis was closely linked with the LCA inputs, using the EOL BESS treatment as a shared basis for both evaluations. This integrated approach allowed for a nuanced analysis, offering a holistic view of the economic implications of sustainability and environmental impact.

3.1 Environmental impact characterization

Within the scope of the provided LCA study, the focus was set on three specific ICs: climate change, eutrophication, freshwater; and resource use, minerals and metals. According to the obtained results, the most significant environmental benefits of BESS EOL treatment are primarily due to the reuse of secondary materials such as aluminium, copper, and steel, which can be effectively recycled and repurposed, extending their lifecycle.

The environmental impacts of the treated components were categorized into battery system, cooling system, fire extinguishing system, and inverters, as depicted in **Figure 3**. Results show significant environmental benefits from recycling and recycling pre-treatment, particularly due to the secondary use of materials like aluminium, steel/iron, copper, and graphite. Consistent with our findings, Kallitsis et al. (2022) reported that secondary aluminium contributes roughly 50% of the environmental benefits in battery recycling. Conversely, secondary steel offers minimal benefits, underscoring the greater environmental advantage of using secondary aluminium in sustainable recycling practices. These benefits outweigh the environmental impacts of transporting recycled materials, using electricity and machinery for recycling, and disposing of non-recycled materials such as waste plastic, electronic scrap, and refrigerant.

The most significant environmental benefits in the climate change IC come from recovering secondary aluminium from batteries, cooling systems, and inverters. In contrast, the benefits of fire extinguisher recycling are mainly due to the recovery of secondary steel. Overall, the environmental benefits outweigh the associated impacts, which mostly stem from the electricity consumption of the machinery and sets, totalling approximately $1.2 \cdot 10^4$ kg CO_2 eq. The benefits are substantial, with a reduction of approximately $-2.54 \cdot 10^4$ kg CO_2 eq. However, it is essential to note that the environmental impacts of recycling pre-treatment

processes for batteries accounts for about one-third of these environmental benefits. Compared to the study by Romare & Dahllöf (2017), which examined various battery recycling pathways, our results show more minor environmental benefits: i.e. -1,023 kg CO_2 eq./1 kg batteries versus -1,536 kg CO_2 eq./1 kg batteries in their study. This discrepancy is due to the Swedish energy mix used in their research, which has significantly lower carbon emissions than the Czech energy mix used in our study. Considering the analysis of the whole BESS unit, the environmental impacts from electricity used in battery recycling account for nearly one-third of the absolute value of the eutrophication, freshwater IC. The energy used to recycle other BESS components is minimal, in comparison. Significant environmental benefits arise from recovering secondary materials, particularly copper from batteries and AC units, steel from fire extinguishers, and aluminium from inverters. The main environmental benefits in this category come from the batteries (-25.36 kg P eq.) and inverter parts (-18.53 kg P eq.). Fire extinguishers contribute negligibly. The AC unit contribution is around 4.91 kg P eq. The environmental impact of the recycling process is minimal regarding resource use, minerals, and metals IC. However, significant benefits arise from recovering secondary metals such as copper, brass, steel, and aluminium. Batteries contribute the most to resource use, minerals, and metals IC, mainly due to the recovery of secondary copper, accounting for -4.68 kg Sb eq. The recycling of AC units follows, while the other components combined contribute -0.73 kg Sb eq.

In conclusion, enhancing EOL treatment efficiency and managing resources can improve the environmental benefits of EOL BESS treatment. The Czech Republic's energy sector heavily relies on fossil fuels, particularly coal, which exacerbates impacts. Reducing this reliance and transitioning to renewable energy would improve recycling outcomes and support broader sustainability goals. This analysis highlights the vital role of EOL treatment in reusing materials. By recovering aluminium, copper, and steel, recycling extends material lifecycles, reduces virgin material demand, conserves resources, and lowers carbon footprints. Ultimately, the environmental benefits of recycling far exceed its impacts, highlighting its essential role in sustainable resource management and environmental protection.

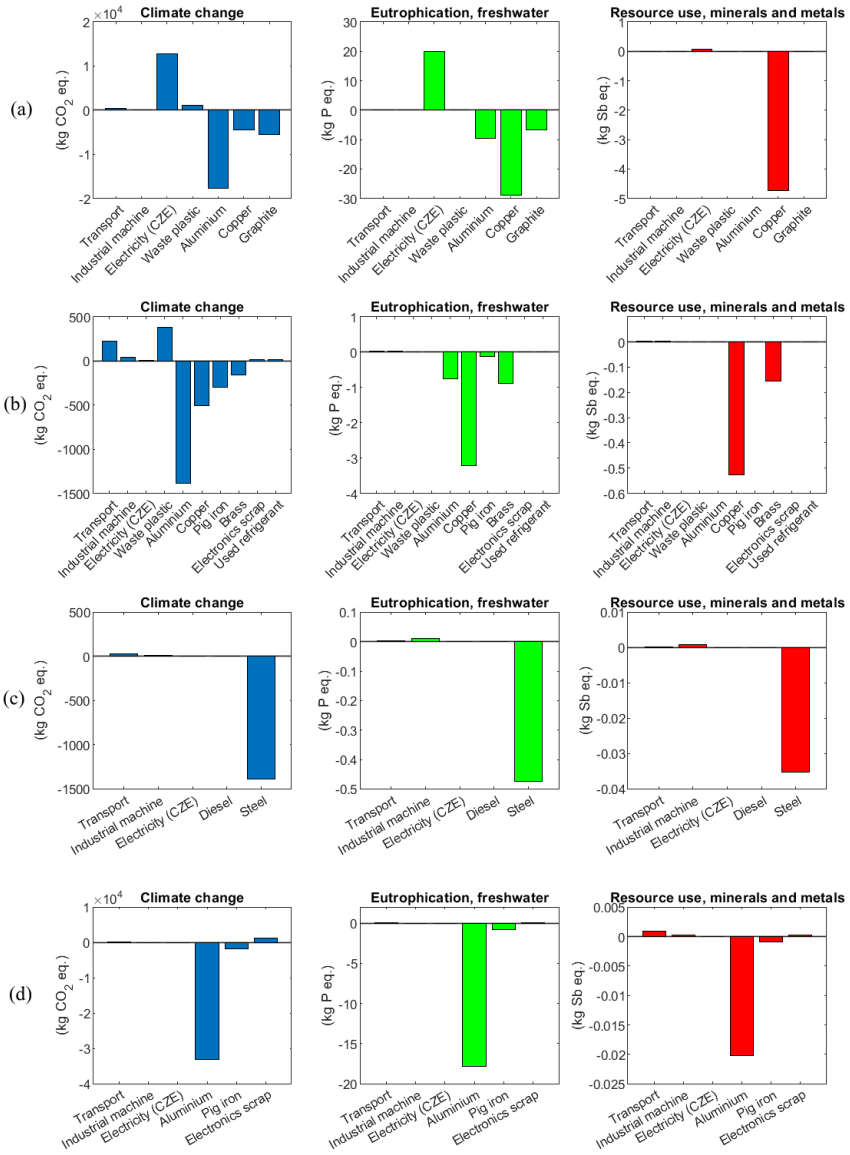

Figure 3. Studied environmental category (climate change, eutrophication, freshwater, resource use, minerals, and metals) results for end-of-life treatment of (a) battery system, (b) cooling system, (c) extinguishing system, and (d) inverters. Numerical data supporting the provided results are included in the supplementary materials.

3.2 Economic viability analysis

The economic analysis results depend on the revenues from recycled materials and recycling costs. Material recovery relies on process efficiency and material inputs based on cathode composition and battery design. The process influences machinery, labour, energy, and transport costs, with potential tax obligations varying by state. This study focused on NMC 6.5:2:1.5 LIB pouch cells, achieving 100% recovery efficiency for metals and graphite in the baseline scenario and different OEE levels covering the operation conditions. Recovered materials include aluminium, brass, cobalt, copper, iron, graphite, manganese, nickel, and steel, with cobalt, nickel, and copper being the most valuable.

Considering the market prices and weights of recycled materials, the analysis reveals that nickel comprises the largest share of potential profits, exceeding 50%. Cobalt follows, trailed by aluminium and copper, constituting more than 90% of the potential profits, as shown in **Figure 4 (a)**. It is essential to highlight the significance of graphite, which represents approximately 6% of the potential profits. However, the strategic importance of graphite lies in the potential for local acquisition because over 60% of graphite is mined in China (Government of Canada, 2024; Statista, 2023) This concern is heightened by the concentration of refining facilities in China, creating dependence on Chinese supplies and vulnerability to their influence. Similarly, the concentration of cobalt in the Democratic Republic of Congo, which produces over half of the global supply, raises concerns about supply chain resilience and geopolitical influence due to heavy reliance on a single source.

The analysis in **Figure 4 (b)** compares potential revenues, costs, and taxation, highlighting energy and labour as major cost drivers. The study indicates machine costs are relatively low, due to the study's focus on specific processes, excluding broader technological support. The cost analysis is based on the EOL treatment of one BESS, with fixed costs tied to work shifts and assuming full-capacity operation. Deviations from total capacity will increase per-BESS recycling costs.

In this case study, assuming 100% efficiency in metal and graphite recovery and an 85% OEE, potential revenues are projected to be €105,000. With current prices for recycled materials, estimated costs are €36,000, including about half for taxes. Thus, recycling BESS under these conditions could yield a profit of around €69,000. The influence of OEE and potential imperfections in the recycling process, which reduces the efficiency of recovering valuable materials from the black mass, is graphically depicted in **Figure 4 (c)**. A decrease in OEE leads to higher costs, while lower recycling efficiency reduces potential revenues. Additionally, as revenues decline and costs rise, potential tax obligations decrease slightly, partially offsetting the negative impact on projected profits.

This focused BESS economic analysis highlights the clear financial benefits of recycling. There is substantial potential to offset the costs of integrating additional technologies and processes into the recycling operation. While profits may decrease when these extra costs are considered, profitability remains promising in the context of EOL treatment. Additionally, local production of raw materials can reduce import reliance, enhance supply chain security, and lower costs and environmental impacts associated with transporting materials from abroad.

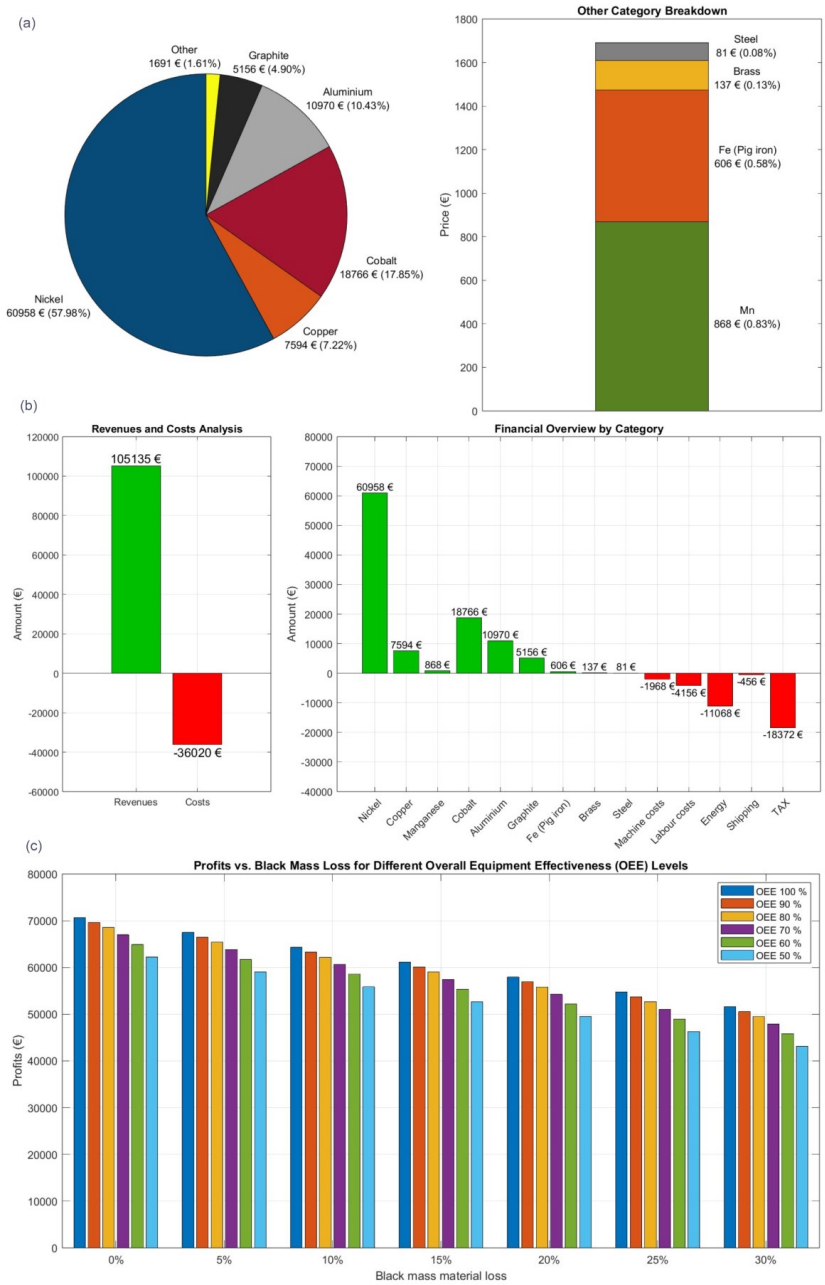

Figure 4. Economic analysis results of end-of-life treatment of battery energy storage system: (a) value of individual materials of system components, (b) revenues and costs analysis, (c) illustration of profits vs. black mass loss for different overall equipment effectiveness levels.

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4. Conclusion

This study evaluated the environmental and economic impacts of EOL treatment for BESS based on LIBs. Focusing on a 2.8 MWh/2.5 MW BESS, the study covers the recycling pre-treatment of batteries, recycling treatment of cooling systems, extinguishing systems, inverters, and the reuse of containers and substations in new BESS installations. The environmental assessment employs a gate-to-gate LCA to measure impacts on three critical ICs: climate change, freshwater eutrophication, and resource use, minerals and metals.

The results indicate significant environmental benefits derived from recycling and pre-treatment processes, primarily due to the secondary use of materials like aluminium, copper, and steel. Notably, secondary aluminium contributes approximately 50% of the environmental benefits in battery recycling, while secondary steel offers minimal advantages. The most significant environmental benefits in the climate change IC arise from recovering secondary aluminium from batteries, cooling systems, and inverters. In contrast, the recycling of fire extinguishers mainly yields benefits from secondary steel recovery. Overall, the environmental advantages of recycling outweigh the environmental impacts associated with the electricity consumption of the machinery used, which totals approximately $1.2 \cdot 10^4$ kg CO_2 eq. The environmental impacts of recycling pre-treatment processes for batteries accounts for about one-third of the observed benefits.

In terms of economic analysis, the study reveals that nickel comprises the largest share of potential profits, exceeding 50%, followed by cobalt, aluminium, and copper, which collectively constitute more than 90% of potential profits. The analysis further emphasizes the strategic importance of graphite, which represents approximately 6% of potential profits but is also critically dependent on local sourcing due to its predominant production in China. The study's findings underscore the potential value of BESS containers at EOL, which can significantly influence investment decisions by offsetting market risks. The documented return from EOL processing of BESS containers can lower investment risks and foster a transition to greener energy. Local production of raw materials further enhances economic benefits by reducing import reliance, improving supply chain security, and lowering costs.

Though based in the Czech Republic, the methodology is applicable globally. In summary, BESS recycling offers triple benefits: environmental improvement through material reuse, reducing reliance on import, and economic gains from local production. Transitioning to renewable energy sources for recycling would further optimize these benefits, aligning with sustainability objectives and ensuring a more efficient and eco-friendly approach to BESS EOL treatment.

CRediT authorship contribution statement

Anna Pražanová: Resources, Methodology, Conceptualization, Writing – Original draft, Project administration.
Zbyněk Plachý: Formal analysis, Data curation, Visualization, Investigation.
Michael Fridrich: Formal analysis, Data curation, Visualization, Investigation.
David Havlíček: Resources, Methodology, Conceptualization.
Jan Weinzettel: Supervision, Writing – Review & Editing.
Daniel-Ioan Stroe: Supervision, Writing – Review & Editing.
Vaclav Knap: Supervision, Writing – Review & Editing, Funding acquisition, Project administration.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Data will be made available on request.

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A method of discharging a battery cell

Field of technology

The present invention relates to a method of discharging battery cells for the purpose of waste and recycling pre-treatment thereof.

Prior art

In the prior art, electrochemical and non-electrochemical processes for discharging (waste) battery cells are known. In electrochemical processes, the discharge of the cells involves contact of electrode contacts with a conductive liquid medium, typically in the form of salt solutions, and significant damage and corrosion of the cell contacts, which is associated with the release of their contents (i. e. packing materials, sleeves, contact materials and cell materials) into the entire volume of the bath and its complete contamination. Toxic components of the electrolytes are also released into the bath, which react with the humidity in the air and evaporate into the air as toxic gases. When implementing these processes, it is therefore necessary to ensure constant ventilation or closed atmospheres, which increase the investment cost of the entire solution. In addition, the application of these conventional electrochemical methods affects the overall quality and quantity of the final product, the black mass (finely ground fraction of the battery cell electrode materials that are rich in rare or important metals). Quality is reduced by corrosion of the materials and contamination of the battery component structures from chemical compounds in the bath solutions, usually salt-based. The blackmass quantity is reduced by releasing the battery cell materials into the bath solution, and for its subsequent recovery, it needs to be isolated, e. g. by additional leaching of the bath solution, which leads to a significant increase in cost. The publication by Nan et al. titled „Recovery of metal values from spent lithium-ion batteries with chemical deposition and solvent extraction“ (Journal of Power Sources 152 (2005) 278-284, available at doi:10.1016/j.jpowsour.2005.03.134) describes in Section 3.1 the discharge of a battery cell using a metal powder in water, under mechanical agitation for 30 min and in a stainless steel container. Despite the published functionality, an empirical drawback has been shown to be the high transient resistance between the metal powder particles, which, without mechanical agitation, does not allow sufficient conduction of the electric current and short-circuiting and discharging of the battery.

The publication by Gratz et al. titled „A closed loop process for recycling spent lithium ion batteries“ (Journal of Power Sources 262 (2014) 255-262, available at http://dx.doi.org/10.1016/j.jpowsour.2014.03.126) describes in section 2.2 (step 1) the discharge of a battery cell using stainless steel chips according to the procedure of Nan et al., therefore in water and for 4 h. The stainless steel chips have no defined shape, size or origin.

The final thesis by Nembhard titled „Safe, Sustainable Discharge of Electric Vehicle Batteries as a Pre-treatment Step to Crushing in the Recycling Process“ (available at http://kth.diva-portal.org/smash/get/diva2:1339943/FULLTEXT01.pdf), describes in a first embodiment in Section 2.7.2 the discharge of a battery cell using a metal powder in a salt solution. The disadvantage of using a salt solution is the corrosion of the electrode contacts of the battery cell, which increases the risk of electrolyte leakage. In another embodiment, Nembhard describes in Sections 3.5, 3.5.1, 4.4 and 4.4.2 the discharging of a battery cell using metal staples (specifically Office Depot Chrome Staples or Rapid Super Strong 9/12 Staples) in distilled water for 1 hour (cell voltage drop to 0.5 V). The staples have a defined shape, an area-to-volume ratio typically less than 2 mm^{-1}, and a thickness typically greater than 0.5 mm. The disadvantages of staples, according to the thesis in Section 4.4.2 and Figure 21, are their overburning, rusting, rust deposition on the battery cell and the unsustainability and impracticality of using staples for this purpose. In addition, overburning and rust formation increases resistance, reduces conductivity, and thus complicates the discharge process.

The European patent application EP 4020663 A1 describes a method of discharging a battery cell by means of an electrically conductive bulk material (e. g. stainless steel chips or staples) in an electrically conductive liquid (e. g. an aqueous salt solution). The stainless steel chips are specified in Example 3 of said document to a substantially conical shape with a diameter of approximately 10 mm, in combination with a 10% aqueous Na_2CO_3 solution. The disadvantage of using a salt solution is the corrosion of the electrode contacts of the battery cell, which increases the risk of electrolyte leakage.

Other methods of discharging battery cells are described in the international patent application WO 2023106416 A1 and in the publications Neha Garg et al., Sustainable Energy Fuels, The Royal Society of Chemistry, 28 May 2024, Vol. 8, p. 2777-2788 titled „Enhanced electrochemical discharge of Li-ion batteries for safe recycling“; Dawei Yu et al, Minerals Engineering, Nov. 1, 2021, Vol. 173, p. 107218 titled „Pretreatment options

for the recycling of spent lithium-ion batteries: A comprehensive review“; and Lin Peng Yao et al., Journal of Cleaner Production, Feb. 1, 2020, Vol. 245, p. 118820 titled „An environmentally friendly discharge technology to pretreat spent lithium-ion batteries“.

Therefore, a need arises in the prior art and the object of the invention is to provide a safe, sustainable and fast non-electrochemical method of discharging a battery cell.

Summary of the invention

Said object is achieved by a method of discharging a battery cell, comprising mixing the battery cell, a liquid coolant and metal fragments such that electrode contacts of the battery cell are immersed in the liquid coolant, wherein the metal fragments have an area-to-volume ratio in the range of 2 to 11 mm^{-1} and a thickness in the range of 0.1 to 0.5 mm. The term „metal fragment“ means a metal chip, swarf or shavings and not a particle or clumps of metal powder.

The underlying idea of the method according to the present invention is that the battery cells are discharged by a short-circuit current generated by connecting the electrode contacts by means of the metal fragments in the liquid coolant. During the discharge process, the voltage across the cell contacts drops very rapidly, with typical discharges below 1 V within 30 min, preferably below 0.5 V within 15 min, for cells in a transport state with ~30% SOC (e. g. between 25 and 35% SOC). Complete discharge and a drop in the total battery cell voltage (i. e. voltage after regeneration for at least 24 h) below 1 V occurs when cells at the end of their lifetime are discharged for at least 5 h, fully charged cells (100% SOC) for at least 18 h, preferably for cells in a transport state (~30% SOC) for at least 15 h. This voltage value is safe for further recycling pre-processing steps, including crushing and grinding steps, and completely eliminates the potential risk of ignition, short-circuiting or sparking.

The term „voltage across electrode contacts of a battery cell“ means the effective voltage across the contacts prior to regeneration of the cell, i. e. prior to charge distribution across the cell. The term „total battery cell voltage“ means the total voltage across the contacts after regeneration of the cell, i. e. after charge distribution across the cell. Regeneration means leaving the cell for a minimum of 24 hours for the voltage to settle. SOC means state of charge.

Discharging below the liquid coolant level brings increased safety to the entire process, eliminating the risk of thermal runaway, sparking or burning of metal fragments. The battery cells can be discharged in large quantities, there is no need to discharge each battery cell individually.

Metal fragments that are too large (i. e. area-to-volume ratio less than 2 mm^{-1} and thickness greater than 0.5 mm) may not make contact between themselves and the electrode contacts of the battery cell to form a conductive path. Too small and irregularly shaped metal fragments (i. e. area-to-volume ratio greater than 11 mm^{-1} and thickness less than 0.1 mm, e. g. metal filings, which are spiral-shaped with sharp edges, rounded parts, hooks, beaks, etc.) can on the other hand get stuck one with another and form clusters that also prevent contact with the electrode contacts of the battery cell, wherein too small a size of metal fragments leads to their rapid overburning by the high short-circuit current of the battery during the resulting electric arc, which leads to a significant reduction in conductivity and thus in an increase in discharge time.

In a preferred embodiment, the metal fragments have an area-to-volume ratio greater than 2.2 mm^{-1}, preferably in the range of 4 to 10 mm^{-1} (e. g. 5 to 8 mm^{-1}), and a thickness of less than 0.45 mm, preferably in the range of 0.1 to 0.25 mm (e. g. 0.1 to 0.2 mm).

In a preferred embodiment, the metal fragments are waste products of subtractive manufacturing processes, in particular machining, cutting, turning or milling.

In a preferred embodiment, the liquid coolant is water (e. g. drinking water, utility water, demineralised water, distilled water, industrial water), mineral oil (e. g. transformer oil), or glycerol, or mixtures thereof (in any ratio), wherein the liquid coolant does not have a corrosive effect on the electrode contacts (unlike e. g. salt solutions).

In a preferred embodiment, the conductivity of the metal fragments is in the range of 0.5 to 60 MS/m, i. e. the metal fragment may be a common metal material such as iron, steel, stainless steel, aluminum or copper.

Brief description of the drawings

Figure 1 schematically shows a container with a liquid coolant, a battery cell and metal fragments.

Figures 2A to 2C show images of metal fragments according to the present invention (metal swarf from machining).

Figures 3A to 3E show images of unsuitable metal fragments (metal filings from cutting).

Figure 4 shows the cell discharge process as a dependence of the voltage across the electrode contacts of the battery cell (SOC~ 30%) on the discharge time in different liquid coolants.

Figure 5 shows the cell regeneration process as a dependence of the total battery cell voltage on the regeneration time in different liquid coolants.

Examples

Example 1

Figure 1 schematically shows a container **1** in which a battery cell **2**, a liquid coolant **3** and metal fragments **4** are mixed. The electrode contacts of the battery cell **2** are immersed in the liquid coolant **3**. A preferred mixing sequence may be: pouring the metal fragments **4** into the container **1**, inserting the battery cell **2**, pouring in more metal fragments **4** and pouring in the liquid coolant **3** (in this case no mechanical mixing is necessary to form a conductive contact of the electrode contacts). Another preferred mixing sequence may be: pouring the liquid coolant **3** into the container **1**, inserting the battery cell **2** and pouring in the metal fragments **4**. The container **1** is preferably made of an electrically non-conductive material (e. g. glass, plastic).

Figures 2A-2C show suitable metal fragments in the form of metal swarf obtained by machining. Such metal swarf is flat, slightly curved in shape with substantially straight edges, such as that shown in Figure 2C, having a thickness t of 68 to 219 μm (average value 143 μm), a width of 1.466 mm, and an area S to volume V ratio of 6.99 mm^{-1} (see first calculation below), and satisfies the condition of an area-to-volume ratio ranging from 2 to 11 mm^{-1} and a thickness ranging from 0.1 to 0.5 mm. For said swarf, the discharge is rapid (after 15 min of discharge, a drop below 0.5 V, followed by recovery after 24 h to below 1 V), see Example 3.

Figures 3A-3E show unsuitable metal fragments in the form of metal filings obtained by cutting. These metal filings are spiral-shaped, strongly curved in shape with sharp, uneven edges and hooked or beaked irregularities etc., e. g. in Figure 3E, having a thickness t of 83 to 95 μm (average value 89 μm) and an area S to volume V ratio of 11.24 mm^{-1}, i. e. dimensionally smaller than the swarf of Figs. 2A-2C, thus not meeting the condition of an area-to-volume ratio in the range of 2 to 11 mm^{-1} (see second

calculation below) and a thickness in the range of 0.1 to 0.5 mm. Clusters of metal fragments are visible in Figs. 3C and 3D, which is a consequence of their shape. For these filings, the discharge is more gradual (dropping below 0.5 V after 25 h of discharge, with subsequent recovery to 0.7 V after 24 h).

$$1.\, Area - to - volume\ ratio_{(machining)} = \frac{S}{V} = \frac{S}{S \cdot t} = \frac{1}{t} = \frac{1}{\frac{0{,}068+0{,}219}{2}} = \frac{1}{0{,}143} = 6{,}99\ mm^{-1}$$

$$2.\, Area - to - volume\ ratio_{(cutting)} = \frac{S}{V} = \frac{S}{S \cdot t} = \frac{1}{t} = \frac{1}{\frac{0{,}083+0{,}095}{2}} = \frac{1}{0{,}089} = 11{,}24\ mm^{-1}$$

Example 2

Further suitable metal fragments in the form of metallic swarf obtained by machining are flat, slightly curved in shape with substantially straight edges, having an average thickness *t* of 435 μm and an area *S* to volume *V* ratio of 2.30 mm^{-1} (see third calculation below), or having an average thickness *t* of 108 μm and an area *S* to volume *V* ratio of 9.26 mm^{-1} (see fourth calculation below). These fragments satisfy the condition of an area-to-volume ratio between 2 and 11 mm^{-1} and a thickness between 0.1 and 0.5 mm. For said swarf, the discharge is rapid (dropping below 0.5 V after 15 min of discharge, followed by recovery below 1 V after 24 h).

$$3.\, Area - to - volume\ ratio_{(machining)} = \frac{S}{V} = \frac{S}{S \cdot t} = \frac{1}{t} = \frac{1}{0{,}435} = 2{,}30\ mm^{-1}$$

$$4.\, Area - to - volume\ ratio_{(cutting)} = \frac{S}{V} = \frac{S}{S \cdot t} = \frac{1}{t} = \frac{1}{0{,}108} = 9{,}26\ mm^{-1}$$

Example 3

Figure 4 shows an example of a dependence of the voltage across the electrode contacts of the battery cell on the discharge time in a liquid coolant - demineralised water (red), drinking water (blue, purple) and transformer oil (green). The voltage across the electrode contacts of the battery cell drops below 0.5 V in less than 10 min for demineralised water and in less than 15 min for drinking (tap) water and transformer oil. In all cases, the voltage across the electrode contacts of the battery cell drops to 0 V after 50 min.

Figure 5 shows a dependence of the total battery cell voltage on the regeneration time after discharge in a liquid coolant - drinking water (blue) and transformer oil (green). In the case of drinking (tap) water, the total battery cell voltage reaches 0.5 V after regeneration for approximately 16 hours. For transformer oil, the total battery cell voltage reaches 0.4 V after regeneration for approximately 14 hours. Since the effective voltage across the electrode contacts of the battery cell in Fig. 4 has dropped to 0 V, the curves in Fig. 5 have an increasing waveform, i. e. the battery cell regenerates and the charge within the cell equalises.

Industrial applicability

The above-described method can be used in waste and recycling pre-processing of battery cells.

List of reference signs

1 container
2 battery cell
3 liquid coolant
4 metal fragment

CLAIMS

1. A method of discharging a battery cell (2), comprising mixing the battery cell (2), a liquid coolant (3) and metal fragments (4) such that electrode contacts of the battery cell (2) are immersed in the liquid coolant (3), **characterised in that** the metal fragments (4) have an area-to-volume ratio in the range of 2 to 11 mm^{-1} and a thickness in the range of 0.1 to 0.5 mm.

2. The method according to claim 1, **characterised in that** the metal fragments (4) have an area-to-volume ratio greater than 2.2 mm^{-1} and a thickness of less than 0.45 mm.

3. The method according to claim 2, **characterised in that** the metal fragments (4) have an area-to-volume ratio in the range of 4 to 10 mm^{-1} and a thickness in the range of 0.1 to 0.25 mm.

4. The method according to any one of the preceding claims, **characterised in that** the metal fragments (4) are waste products of subtractive manufacturing processes.

5. The method according to claim 4, **characterised in that** the metal fragments (4) are chips or swarf obtained by machining, cutting, turning or milling.

6. The method according to any one of the preceding claims, **characterised in that** the liquid coolant (3) is water, mineral oil or glycerol, or mixtures thereof.

7. The method according to any one of the preceding claims, **characterised in that** the conductivity of the metal fragments (4) is in the range of 0.5 to 60 MS/m.

A. Pražanová, Z. Plachý, and V. Knap, “Způsob vybíjení bateriového článku” [P1], Czech patent, CZ2024247A3, 2024, filed also as European patent application EP25157404.2 on 12 Feb. 2025.

ABSTRACT

The present invention relates to a method of discharging a battery cell (2), comprising mixing the battery cell (2), a liquid coolant (3) and metal fragments (4) such that electrode contacts of the battery cell (2) are immersed in the liquid coolant (3). The metal fragments (4) have an area-to-volume ratio in the range of 2 to 11 mm^{-1} and a thickness in the range of 0.1 to 0.5 mm.

Figure for abstract: Fig. 1

A. Pražanová, Z. Plachý, and V. Knap, “Způsob vybíjení bateriového článku” [P1], Czech patent, CZ2024247A3, 2024, filed also as European patent application EP25157404.2 on 12 Feb. 2025.

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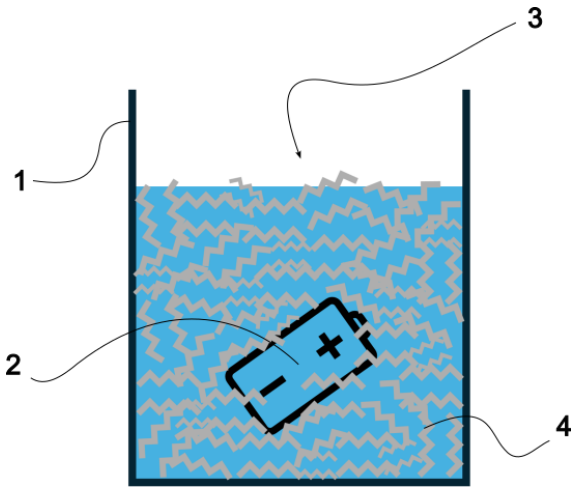

Fig. 1

Fig. 2A

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Fig. 2B

13

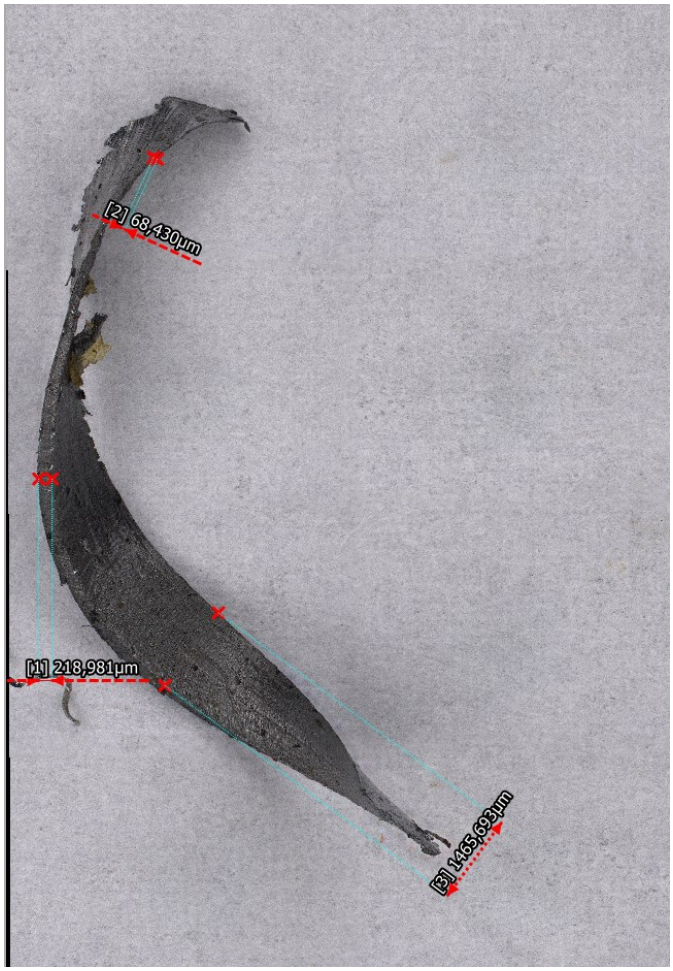

Fig. 2C

14

Fig. 3A

Fig. 3B

5

15

Fig. 3C

Fig. 3D

5

16

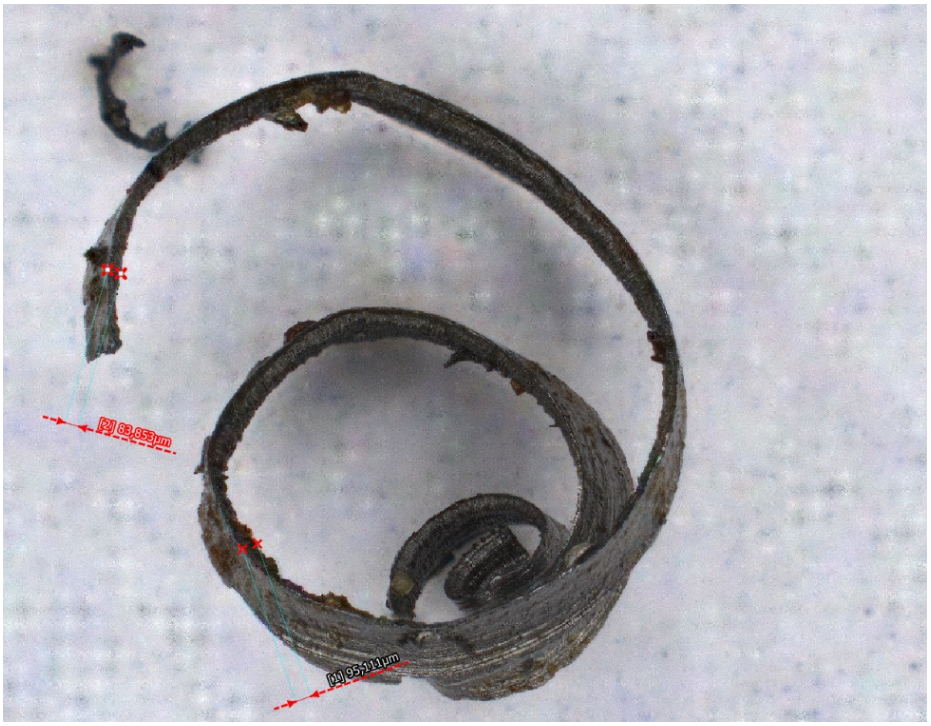

Fig. 3E

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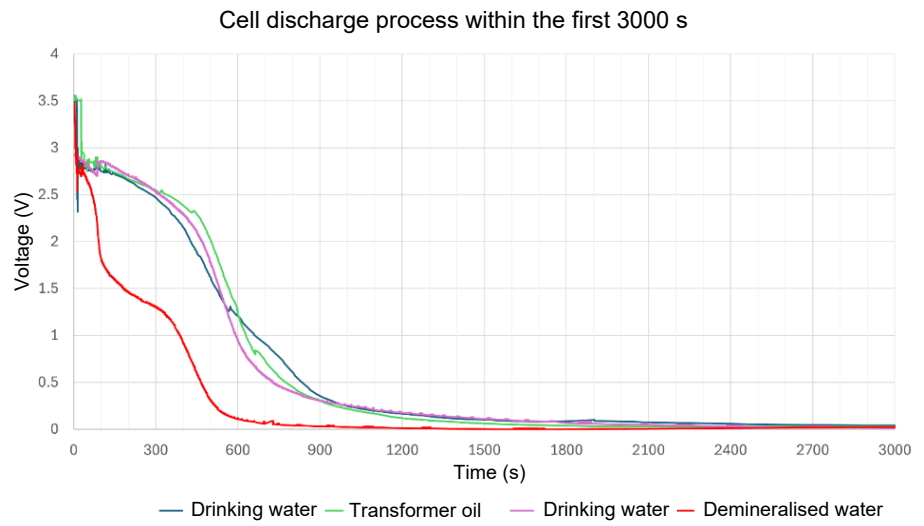
Cell discharge process within the first 3000 s
Voltage (V)
Time (s)
Drinking water
Transformer oil
Drinking water
Demineralised water

Fig. 4

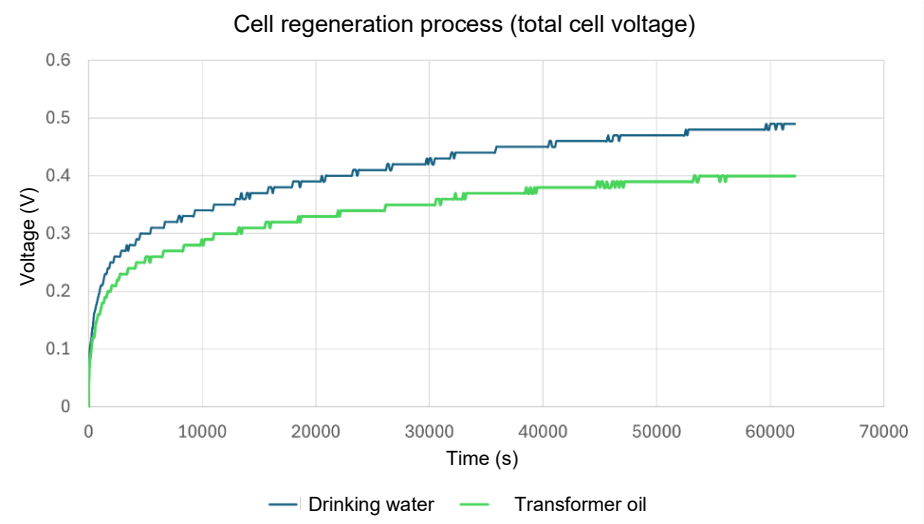
Cell regeneration process (total cell voltage)
Voltage (V)
Time (s)
Drinking water
Transformer oil

Fig. 5

A. Pražanová, *et al.*, "Techno-Environmental Evaluation of Recycling Pretreatment of Cylindrical Lithium-Ion Battery: Discharging via Salt-Based Solution" [C1], © 2023 IEEE. Reprinted, with permission, from IEEE.

Techno-Environmental Evaluation of Recycling Pretreatment of Cylindrical Lithium-Ion Battery: Discharging via Salt-Based Solution

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***Abstract*— The popularity of lithium-ion batteries (LIBs) as crucial power sources has increased in recent years. LIBs represent a perspective technology for recycling because they comprise a high portion of valuable metals, such as nickel, manganese, cobalt, or lithium, and other metals, including aluminium, copper, and iron. Battery discharging represents an essential step in end-of-life (EOL) pretreatment, as it reduces the risk of fire or explosion in further processing. As a simple, quick, and inexpensive technique, an electrochemical discharging process via salt-based solutions is preferred for cylindrical cells. Nevertheless, it is necessary to consider the composition of obtained waste products and the possible environmental risks leading to their safe and non-hazardous EOL processing. This work evaluated discharging efficiency and environmental perspective for cylindrical LIB cells, which were treated using NaCl solution. All battery cells were discharged to the safe voltage limit (0.75 V) within 24 hours. Major organic components, including volatile solvents with high toxic hazards, such as carbonic acid esters, methyl salicylate, and propanoic acid esters, were identified in the waste solutions using gas chromatography with mass spectrometry (GC-MS). Moreover, the metal proportion in the solution was determined using inductively coupled plasma - optical emission spectrometry (ICP-OES) analysis; it is recommended to recover metals from the wastewater before EOL or cleaning treatment.**

Keywords—lithium-ion battery, discharging, recycling, environmental impacts, safety

I. Introduction

Nowadays, the use of lithium-ion batteries (LIBs) is increasing worldwide; they are considered crucial power sources for personal electronic devices, such as smartphones, e-readers, or laptops, as well as for means of electromobility transport, such as e-bikes, e-scooters, or electric vehicles [1].

Moreover, LIBs represent a highly perspective technology for end-of-life (EOL) treatment, especially for recycling. They are typically based on valuable metals, including lithium (Li), cobalt (Co), manganese (Mn), and nickel (Ni) in the active cathode materials, graphite (C) in the anode materials, and aluminium (Al), copper (Co), or iron (Fe) in other components such as current collectors, or casings. However, the battery cells also comprise an electrolyte and other highly volatile and reactive compounds, such as lithium hexafluorophosphate ($LiPF_6$) salt, which can react with water and air humidity and result in toxic products [1], [2].

Battery discharging represents an essential step in EOL pretreatment, as it reduces the risk of fire or explosion in further processing, including crushing. Cylindrical cells are typically discharged using electrochemical processes using the salt solution. They are commonly based on ionic solutes with sodium (Na^+), potassium (K^+), or ammonium ($NH4^+$) cations, and mainly following solutes: sodium chloride (NaCl), sodium hydrogen sulphate ($NaHSO_4$), sodium hydroxide (NaOH), potassium chloride (KCl), potassium bromide (KBr), or ammonium sulphate ($(NH_4)_2SO_4$). The immersion in the solution leads to controlled short-circuiting, where the electrolysis eliminates the battery charge and leads to its discharging [3], [4].

Nevertheless, it is necessary to consider the risks of introducing this simple, quick, and inexpensive solution, typically used as a discharging step during the EOL processing of cylindrical LIB cells. It concerns the composition of the waste solutions (wastewater), the evaluation of their toxicity, and potential hazards after discharging to the required voltage limit [3], [5]. Additionally, it is advisable to also focus on the obtained waste product and its potential in terms of recovery of valuable metals before EOL or cleaning treatment.

This work evaluated the process efficiency and environmental impacts of discharging cylindrical LIB cells in NaCl solution, which was chosen as a highly preferred one for use in a recycling line due to its low costs and high availability. Six types of LIB cells, in nickel-manganese-cobalt (NMC) or lithium-iron-phosphate (LFP) chemistry, 1.5–5 Ah capacity, and their original shipped or fully charged state, were discharged using 5 or 10 wt. % NaCl solution under the voltage limit of 0.75 V within 24 hours. This voltage was in this work considered as safe for further treatment within the recycling plant. The obtained solutions were further analysed for their toxicity and potential risks using gas chromatography with mass spectrometry (GC-MS). In addition, an inductively coupled plasma - optical emission spectrometry (ICP-OES) analysis for the metals proportion in wastewater was performed. The solutions contain a significant proportion of carbonic acid esters, methyl salicylate, and propanoic acid esters, characterized by high risks that must be considered during the treatment. Moreover, a high metals proportion was identified in the wastewater medium. Therefore, it is advisable to process the waste solutions before their cleaning with a suitable recycling technique (e.g., hydrometallurgy) to recover the selected metals.

II. EXPERIMENTAL

Six LIB cylindrical cells, including nickel-manganese-cobalt (NMC) or lithium-iron-phosphate (LFP) chemistry in the capacity range of 1.5–5 Ah, were discharged using NaCl solution in distilled water. Discharging measurements were performed using continuous measurement by an Arduino platform and manual voltage measurement using a digital multimeter. The cells were discharged below the voltage level of 0.75 V. A detailed analysis of the main hazardous substances using headspace solid-phase microextraction (SPME) extraction coupled to gas chromatography with mass spectrometry (GC-MS) was performed for the obtained waste solutions. Moreover, the metal proportion for solutions was determined using inductively coupled plasma - optical emission spectrometry (ICP-OES).

A. Materials

Selected types of cylindrical LIB cells from five different producers were experimentally discharged in their original shipped state of charge or fully charged state. The specifics of the tested cells, characterizing their producer, chemistry, capacity, and discharge status, are listed in Tab. I.

TABLE I. TESTED LITHIUM-ION BATTERY CELL SPECIFICS.

Producer	*Chemistry*	*Capacity (Ah)*	*Discharge status*
SAMSUNG	NMC	1.5	Shipped state
O'CELL	LFP	1.5	Shipped state
SAMSUNG	NMC	2.6	Shipped state
CHAM	NMC	2.6	Shipped state
BAK	NMC	5	Shipped state
GOOWEI	LFP	5	Shipped and fully charged state

B. Methods

The cells were first discharged by immersion into the NaCl solution for 24 hours. After evaluating the discharging process efficiency, the GC-MS and ICP-OES analysis for the waste solutions was performed regarding safety, possible risks, and metal recovery potential during the recycling plant operation.

1) Discharging procedure

All cell types were discharged in a 5 wt. % NaCl in distilled water solution in their original shipped state of charge. In addition, the GOOWEI LFP 5 Ah cells were discharged in their fully charged state using 5 wt. % and 10 wt. % NaCl solution.

Continuous measurement using an Arduino UNO R3 CH340 platform with a 30-minute measurement step (in solution) and manual voltage measurement using Voltcraft VC185STRMS digital multimeter (out of solution) was performed to monitor the discharging process. For Arduino-measured battery cells, the cell electrode contacts were treated with lead-free solder; the extended electrode terminals were soldered. The electrode contacts of the manually measured cells were not treated in any way. Each discharging procedure lasted 24 hours; to evaluate the process, a discharging voltage limit of 0.75 V was set, which was considered safe for further processing in the recycling plant including crushing.

2) Hazards, risks, and material recovery evaluation

The evaluation of potential risk and hazardous substances was determined based on the analysis using headspace SPME extraction coupled to GC-MS for the waste solutions. A Thermo Fisher Scientific Trace 1310 GC coupled to an ISQ 7000 mass spectrometer identified volatile compounds released out of the discharging solution. An autosampler TriPlus RSH used an SPME arrow (CWR-DVB-PDMS) extraction of organic compounds present in the sample to achieve low detection limits. 1 ml of sample was incubated at 80 °C for 30 minutes, followed by 20 min extraction. GC separation ran on semi-standard non-polar column TG-5SILMS (30 m; 0.25 mm; 0.25 µm) with helium as a mobile phase (1.2 ml/min). The oven program started at 40 °C held for 1 minute, followed by a 5 °C/min gradient up to 280 °C. A single quadrupole mass spectrometer was set to full scan acquisition with a 5 Hz scan rate. Spectral identification by MS Search 2.3 with NIST17 preceded blank subtraction and AMDIS deconvolution.

The potential to recover suitable metals from wastewater was evaluated by performing ICP-OES measurements. For quantitation of each metal in the discharging solution containing electrolyte, Agilent 5900 was set to monitor emission bands of Al (396.152 nm), Co (230.786 nm), Cu (324.754 nm), Fe (258.522 nm), Li (670.783 nm), Mn (259.372 nm) and Ni (231.604 nm) in synchronous vertical dual view mode to extend the linear range and improve accuracy. The samples were centrifuged and diluted 100x into 2% HNO_3, 10x if below LOQ.

III. RESULTS AND DISCUSSION

This work was focused on evaluating the process and efficiency of discharging cylindrical LIB cells in NaCl solution. The obtained waste solutions were analysed in terms of their toxicity and risks that would pose a danger during the operation of the recycling line, as well as from the perspective of possible metal recovery.

A. Discharging via NaCl-based sollution

The battery cells begin to discharge electrochemically immediately after their wholly immersed in the solution; bubbles are released from the electrodes and rise to the surface, as shown in Fig. 1 (a). During bubbling (without damage to the electrode contacts), a slow discharge occurs, characterized by a slight change in the resulting voltage.

(a) (b)

Fig. 1. Example of discharging SAMSUNG NMC 1.5 Ah in 5 wt. % NaCl sollution, (a) the process beginning - immediately after immersion; (b) the process end - after 24 hours of discharging.

This process irreversibly damages the surface of the contacts and leads to their complete failure and leakage of the contents of the cells into the discharge solution. A cell with released contents in the discharge solution (with damage electrode contacts) is shown in Fig. 1 (b).

Leakage of battery cell content characterized by a voltage drop is evident by the Arduino measurement, shown in Fig. 2. Moreover, these results also show that mixing the NaCl solution with the electrolyte and the proportion of active materials does not stop the process and the cell still discharges.

For all cells, except SAMSUNG NMC 1.5 Ah and GOOWEI LFP 5 Ah, a drastic voltage drop was recorded during the 30-minute measurement step. A more detailed description of the process, characterizing whether the cell leaks out immediately or gradually, would be achieved by choosing a finer measurement step. The voltage dropped gradually for SAMSUNG NMC 1.5 Ah and GOOWEI LFP 5 Ah cells without significant fluctuations. This can be caused, for example, by a different design or the composition of the electrode contacts.

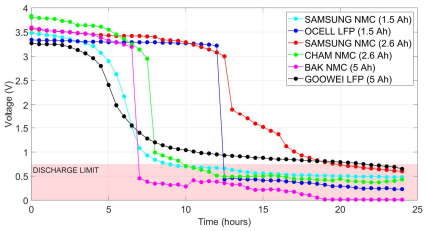
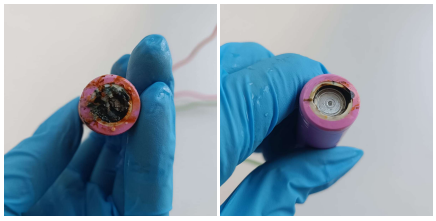


Fig. 2. Example of discharging cylindrical cells in a 5 wt. % NaCl solution measured by Arduino UNO R3 CH340.

Cell electrode contacts are irreversibly damaged during discharge; the final form is shown in Fig. 3, where (a) captures the cell immediately after it is pulling out from the solution and (b) after its purification. The process of destruction of the contacts was slowed down if a layer of solder was applied to the electrodes. A comparison discharging process of treated (with a layer of lead-free solder) cells measured by Arduino and untreated cells measured manually is illustrated in Fig. 4.



(a) (b)

Fig. 3. Example of battery cell contact destruction: (a) immediately after it is pulling out from solution; (b) after purification.

Less drastic voltage drops were recorded for manually measured cells. It is probably caused by the cells pulled out from the solution during each measurement and the contacts cleaning with distilled water. This technique could slow down contacts destruction. Since the cells were not measured manually for the entire 24 hours, the obtained values were fitted with the Smoothing Splines function of the MATLAB Curve fitter software for a better illustration.

All experimentally discharged cells were discharged below the 0.75 V voltage limit within 24 hours. It is clear from the results that the discharge is affected by both the capacity and the material composition of the cell. Discharge can be slowed if the cell electrode contacts are treated, e.g., by soldering; however, both nontreated and treated cells were successfully discharged within the time horizon.

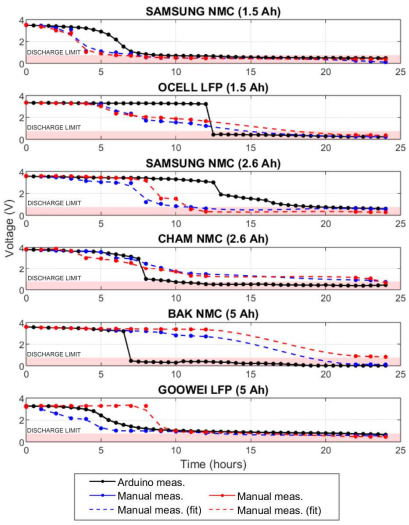


Fig. 4. NaCl solution discharging of LIB cylindrical cells, comparison of anual and Arduino UNO R3 CH340 measurement technique.

In addition, the discharge of the GOOWEI LFP 5 Ah cells in its shipped state of charge was compared with the fully charged state, and when discharging 5 wt. % or 10 wt. % NaCl solution. Charging the cell and increasing the concentration of NaCl in the solution led to an increase in the rate of the discharge process. However, the discharge method did not affect the resulting voltage, which varied from 0.65–0.73 V for all three cells after 22. hour of measurement. A comparison and discharge process for individual cells is shown in Fig. 5.

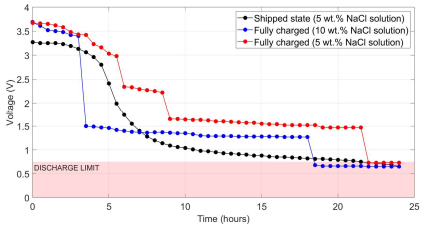


Fig. 5. NaCl solution discharging of GOOWEI LFP 5 Ah.

B. Safety and recovery aspects for recycling plant

The waste solutions were analysed using headspace SPME extraction coupled with GC-MS. Obtained chromatograms are illustrated in Fig. 6. A significant ratio of volatile carbonic acid esters (ethyl, dimethyl, diethyl, methyl ethyl) was determined. These compounds are primarily used as solvents of electrolyte salts. Longer alkyl chains were observed in most samples and esters of other organic acids, e.g., propanoic or acetic. Another component common to all samples was di-tert-butylphenol, a frequently used antioxidant, possible release from a polymer cover of the cell. Scarce occurrence of aliphatic hydrocarbons may be explained alike. The LFP cells contain an overall wider range of organic compounds with a higher ratio of long-chained carbonates.

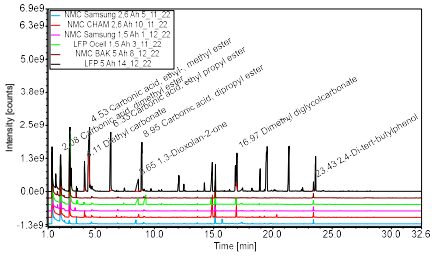


Fig. 6. GC-MS chromatograms of waste solutions with major components.

These volatile solvents induce the main hazards considering larger-scale processing, e.g., in the recycling plant. They pose risks such as high flammability and long-lasting toxicity, which is dangerous for humans and the environment. Even in smaller-scale conditions, they can lead to eye/skin/respiratory irritations. The risks for selected solvents are listed in Tab. II, which are based on GHS hazard statements, lethal dose (LD50) and no observable adverse effect (NOAEL).

TABLE II. TOXICITY ASPECTS OF IDENTIFIED SOLVENTS.

Compound	Structure	Hazard statement	LD50 oral	LD50 skin	LD50 intraperitoneal	NOAEL oral	NOAEL lungs
Dimethyl carbonate		H225	6.4-12.8 g/kg	> 10 ml/kg	0.8-1.6 g/kg	7 mg/kg/day	>1000 ppm
Ethyl carbonate		H318 H319 H302 H373 H315	10 g/kg	>3 g/kg	N/A	1250 mg/kg/day	25000 ppm
Diethyl carbonate		H226 H315 H319 H335	>4,5 g/kg	N/A	8,5g/kg	7 mg/kg/day	N/A
Di-tert-butylphenol		H302 H315 H318 H335 H373 H410	200 mg/kg	200 mg/kg	25 mg/kg	20-75 mg/kg/day	N/A

Source: PUBCHEM/ECHA

According to the performed risk analysis of wastewater, the recycling treatment, starting from the aqueous battery cell discharging step, must be carried out under increased safety conditions. The discharging procedure should be conducted in a closed atmosphere without releasing vapours or other potentially dangerous substances.

Furthermore, ICP-OES analysis of the obtained solutions was performed to evaluate their potential use in recovering suitable materials. A significant proportion of both rare and other metals, such as Ni, Co, Li, Mn, Al, Cu, Fe, was determined in the battery cell solutions as is shown in Fig. 7.

A significant proportion of metals is released into the aqueous medium by LIB cell discharging in NaCl solution. Therefore, considering the scenario of the recycling line, this waste product should be treated, for example, within hydrometallurgical methods leading to full selected material recovery.

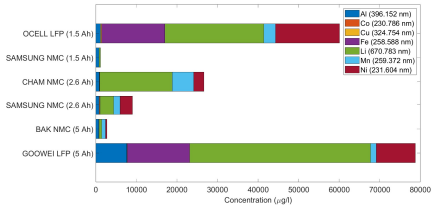


Fig. 7. ICP-OES analysis results: metals proportion in wastewater obtained from 5 wt. % NaCl solution used for LIB cell discharging.

IV. CONCLUSIONS

This work evaluated the efficiency and environmental effects of discharging cylindrical LIB cells in 5 or 10 wt. % NaCl solution in distilled water. Within 24 hours, the cells, both in their original shipped or fully charged state with the capacity of 1.5–5 Ah, were discharged under a voltage of 0.75 V. The obtained waste solutions contain a high proportion of metals and are suitable for further processing within the recycling line, where selected metals will be recovered. The entire process, starting from the discharge step, must be carried out under increased safety conditions, limiting the leakage of toxic compounds based on carbonic acid esters, methyl salicylate, and propanoic acid esters identified in the waste solutions. In the follow-up work, the possibilities of wastewater treatment will be evaluated, including, for example, the use of UV methods or adsorption techniques.

ACKNOWLEDGMENT

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A. Pražanová, *et al.*, "Sustainability Challenges: The Circular Economy Dilemma in Lithium-Ion Battery Cell Electrochemical Discharging Processes" [C2], © 2024 IEEE. Reprinted, with permission, from IEEE.

Sustainability Challenges: The Circular Economy Dilemma in Lithium-Ion Battery Cell Electrochemical Discharging Processes

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***Abstract*—Recycling lithium-ion batteries (LIBs) is crucial for environmental sustainability and resource conservation. However, current recycling procedures, particularly for smaller battery formats, pose several challenges. With the increasing demands of the circular economy for LIB waste treatment, it is essential to identify and address obstacles associated with concurrent processes. Thus, this study focuses on characterizing and examining the dilemmas of electrochemical discharging of cylindrical LIB cells. It primarily examines the quantity and composition of released battery mass from nickel-aluminium-cobalt (NCA) LIB cells using aqueous discharging via solutions of sodium chloride (NaCl), sodium hydroxide (NaOH), and sodium nitrate ($NaNO_3$) within the 5-30 wt. % range. Additionally, the work monitored several procedure parameters, including the voltage profiles during discharging, the extent of battery contacts and casing damage after discharging, the character and material composition of the obtained battery mass, and the composition of the wastewater obtained after separating the solid product from waste solutions. Consequently, it was determined that the industrial implementation of these procedures, including material leakage and disposal, may incur economic losses of up to 1560 USD/tonne due to metal loss.**

Keywords—lithium-ion battery, end-of-life, recycling, sustainability, wastewater

I. Introduction

Considering environmental sustainability and resource conservation, recycling lithium-ion batteries (LIBs) represents an essential end-of-life (EOL) management procedure. Nevertheless, recovering crucial battery elements from battery active materials, current collectors, or casings serves several challenges, from battery discharging to highly efficient material treatment [1]. To mitigate the safety hazards of battery disassembly during EOL treatment procedure, the battery state-of-charge (SOC) should be optimally maintained below 0 %, corresponding to a voltage below 2 V. Therefore, various approaches, such as discharging or needling under an inert and low-temperature environment, have been suggested [2]. However, the economic feasibility of these solutions, particularly for batteries of diverse sizes, such as those in cylindrical cell formats, poses challenges, especially when considering large-scale industrial implementation [3].

At present, two common methods are favoured for discharge recycling pre-treatment [3]. The first involves electrochemical discharge in a conducting solution, entailing the immersion of the battery in a salt-based solution, covering solutes with sodium (Na^+), potassium (K^+), or ammonium (NH_4^+) cations [4]. While this method offers high process efficiency and simplicity, it has the significant drawback of releasing hazardous and highly volatile battery substances into the solution [2], resulting in the generation of toxic wastewater. The second method entails non-electrochemical discharge in conductive powders. Although it avoids the destruction of battery contacts, its implementation, with low efficiency, high complexity, and safety concerns, is presently undergoing optimization efforts [3].

Considering the increasing demands of the circular economy for LIB waste treatment, it becomes crucial to identify and address challenges arising from concurrent processes [5]. This work provides a comprehensive assessment of electrochemical discharging of cylindrical LIB cells. Specifically, it aims to analyse the quantity of released battery mass and evaluate potential cost losses, along with other considerations, that may arise during industrial-scale implementation. In detail, it monitors the discharge process, battery contact destruction effects, and the quantity and composition of the released battery mass across different concentrated solutions of sodium chloride (NaCl), sodium hydroxide (NaOH), and sodium nitrate ($NaNO_3$) within the 5-30 wt. % range. Additionally, the wastewater solutions composition was identified and the economic losses and sustainability implications during the industrial application of these discharge techniques were assessed.

II. Experimental

In electrochemical discharging experiments, this work compared different concentrations ranging from 5 to 30 wt. % of three solutions: NaCl, NaOH, and $NaNO_3$. Moreover, the portions of released battery mass were studied and considered within the context of the circular economy.

A. Materials

In their original shipped state, three types of cylindrical LIB cells from various manufacturers that differ in chemistry, capacity, and dimensions were discharged. Their characteristics are shown in detail in Tab. I.

TABLE I. Specifics of experimentally tested cylindrical lithium-ion battery cells.

Producer	Type	Capacity (mAh)	Dimensions (mm)	Weight (g)
JGNE	LFP	1800	18.20 x 65.20	45
SAMSUNG	NCA	3500	18.75 x 65.25	50
BAK	NMC	5000	21.30 x 70.50	70

B. Methods

The LIB cells, initially at around 3.5 V, underwent a 10-hour discharge by immersion in prepared solutions, with continuous voltage measurement via the Arduino platform. The battery's solid mass was then extracted from the wastewater using a laboratory centrifuge. The dried mass underwent scanning electron microscopy (SEM) analysis, including energy dispersive X-ray spectroscopy (EDS) mapping for composition determination. Wastewater solutions were treated using gas chromatography-mass spectrometry (GC-MS) for toxicity and sustainability characterization.

1) Electrochemical Discharging Procedure

Continuous measurement was conducted using an Arduino UNO R3 CH340 platform with a 2-minute measurement interval step during the discharging process. For the measurement, the battery contacts were initially welded; then the extended terminals were soldered using SAC (tin-silver-copper) 305 solder, comprising 96.5% tin (Sn), 3% silver (Ag), and 0.5% copper (Cu).

2) Treatment of Released Battery Mass

Following a 10-hour discharge, the solid component of the battery mass was obtained from waste solutions through centrifugation at 5000 RPM (Hettich Universal 320 R centrifuge). Subsequently, the solid battery mass fraction was dried at 80 °C for 2-6 hours, depending on the quantity obtained.

3) Scanning Electron Microscopy

Furthermore, the structures were characterized with a scanning electron microscope (SEM), the TESCAN VEGA 3 LMU model, at 20 kV, with a beam intensity of 15 and a working distance of 15 mm. Surface morphologies were recorded in a secondary electron (SE) mode and distribution of elements and different phases in a backscattered electrons (BSE) at a magnification of 200x, 1000x and 2000x. The scanned areas at these magnifications were approximately 0.5 mm², 0.02 mm², and 0.005 mm², respectively. In addition, a micro-X-ray elemental map was generated using energy dispersive spectrometry (EDS). The relative error for each element during SEM-EDS measurements (conducted under high vacuum) was below 1%. Elemental composition measurements were taken twice at each magnification, and the results from all six measurements were subsequently averaged.

4) Gas Chromatography with Mass Spectrometry

The wastewater solutions were analyzed using gas chromatography with mass spectrometry (GC-MS) consisting of a Thermo Fisher Scientific Trace 1310 GC coupled to an ISQ 7000 MS. Solid-phase microextraction (SPME) arrow (CWR-DVB-PDMS) on TriPlus RSH autosampler was used. For analysis, 2 ml of samples were incubated at 80 °C for 30 mins, followed by 20 mins extraction. GC separation ran on semi-standard non-polar column TG-5SILMS (30 m; 0.25 mm; 0.25 µm) with helium as a mobile phase (1.2 ml/min). The oven program started at 40 °C held for 1 minute, followed by a 5 °C/min gradient up to 280 °C. The spectrometer was set to full scan acquisition with a 5 Hz scan rate. Spectral identification was provided by MS Search 2.3 with NIST17.

III. Results and Discussion

This work comprehensively evaluates the electrochemical discharge of cylindrical LIB cells, particularly addressing sustainability in industrially-scale applications. It includes selecting a representative cell type preferred for the following experiments, assessing discharge procedure variations, analysing released battery mass quantity and composition, describing destruction effects, characterization of wastewater solution composition, and discussing sustainability aspects.

A. Comparison of Battery Types During Discharging

At the outset of the experiments, all types of LIB cells were discharged in a commonly widely preferred sodium chloride (NaCl) solution [1], with set concentrations of 5 wt. % and 30 wt. %. The quantity of released battery mass is depicted in Fig. 1.

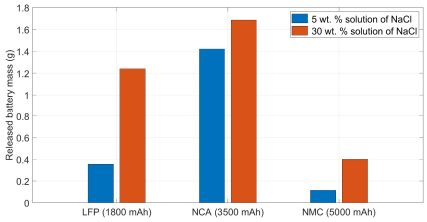

Fig. 1. Released mass from cylindrical battery cells in LFP, NCA, and NMC chemistry via electrochemical discharging using NaCl solutions at 5 and 30 wt. % concentrations.

The NCA LIB cell, with a capacity of 3500 mAh, exhibited the highest proportion of released battery mass. Thus, this cell type was used to represent further assessments and experiments. Controversially, the NMC cell, despite having the largest tested capacity, battery mass, and dimensions, released the most minor proportion of battery mass. As the energy capacity of the cell increases, the reaction becomes more intense, leading to more significant damage to the battery packs [2], [4]. Therefore, a larger battery mass release would be expected. For a comprehensive understanding of the process, it would be essential to provide an analysis of the entire battery structure and the casing material composition for various battery types.

B. Evaluation of Solution Discharging Effect

An illustration of the NCA LIB cells discharge in solutions at concentrations of 5 and 30 wt. % is shown in Fig. 2.

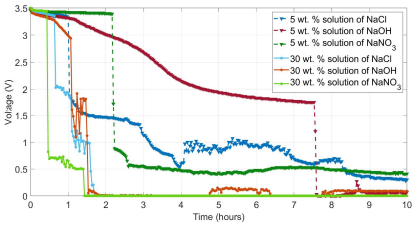

Fig. 2. Example of measured discharge profiles for NCA battery cells, electrochemically discharged via NaCl, NaOH, and $NaNO_3$ solutions at concentrations of 5 wt. % (dashed line) and 30 wt. % (solid line).

For all tested cells, the battery contacts were damaged, releasing the battery mass into the solution. From Fig. 2, it is evident that the concentration significantly influences the discharge rate. For instance, 30 wt.% solutions resulted in a voltage drop to 0 V within 3 hours. Conversely, when discharging 5 wt.% NaCl or $NaNO_3$ solutions, even after 10 hours, the cells maintained a voltage of around 0.5 V, which is deemed safe for further processing, e.g., crushing.

C. *Characterization of Released Battery Mass*

The proportion of released battery mass from NCA LIB cells during discharge in different solutions with concentrations of 5 wt. % and 30 wt. % is provided in Fig. 3.

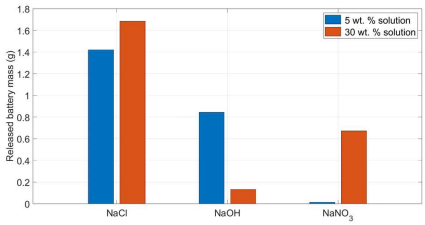

Fig. 3. Released mass from cylindrical NCA battery cells via electrochemical discharging using NaCl, NaOH, and $NaNO_3$ solutions at concentrations of 5 and 30 wt. %.

The highest proportions of material were released during discharge in NaCl solutions, in both tested concentrations. However, in the case of NaOH, more material was released with a lower solution concentration. Discharge in $NaNO_3$ at a concentration of 5 wt. % resulted in the minimum released mass caused only by the destruction of battery contact.

Illustration of obtained waste solutions, battery destruction, and dried released battery mass from the scenario of 30 wt. % concentration discharging is shown in Fig. 4.

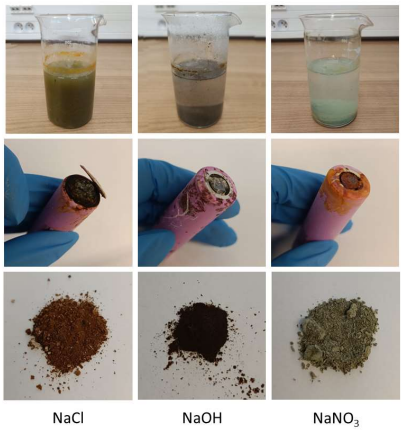

Fig. 4. Example of: top row – aqueous solutions after battery discharge (in 30 wt. % concentration); middle row – battery cell contacts and casing destruction; bottom row – dried released battery mass.

Furthermore, the obtained battery mass in powders was characterized using SEM and EDS mapping, shown in Fig. 5. The mass released from NaCl discharge, ranging from 20 to 70 µm, primarily comprises NaCl residues coupled with battery elements. In contrast, particles from NaOH discharge are fine, typically up to 20 µm. The $NaNO_3$ sample contains material clusters of varying sizes, reaching up to 30 µm.

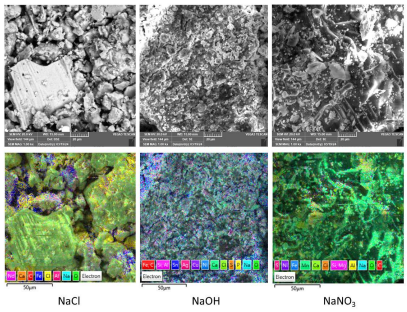

Fig. 5. Material analysis results for battery mass from 30 wt. % discahrging: top row – SEM detail at 1000x magnification; bottom row – EDS mapping showing identified elements.

The comprehensive results of the EDS element maps are further detailed in Tab. II. These results indicate that discharge destroys battery contacts and casings (Cr, Fe, Ca, Cl, Mn, Mg, S, Si), solder (Sn, Ag, Cu), and electrode materials (Ni, Al, C, P). Although cobalt (Co) is present in NCA battery cells, no release in solutions was detected. This is due to its relatively small Co content in only one specific form (electroactive material on the cathode) in the internal structures of the battery and strong covalent bonds in the crystal structures of the materials, which are not affected by the solutions used.

TABLE II. ENERGY DISPERSIVE X-RAY SPECTROSCOPY ELEMENT MAP RESULTS FOR THE RELEASED BATTERY MASS (30 WT. % SOLUTIONS).

Solution	**NaCl**		**NaOH**		**$NaNO_3$**	
Element	***weight (wt. %)***	***σ (wt. %)***	***weight (wt. %)***	***σ (wt. %)***	***weight (wt. %)***	***σ (wt. %)***
Ag			0.53	0.15		
Al	5.57	1.55	2.03	0.69	2.93	1.87
C	8.73	2.10	4.28	1.03	14.85	2.62
Ca	0.32	0.04	1.67	0.62	4.10	1.60
Cl	32.15	3.54	0.23	0.14	0.95	0.54
Cr	-	-	-	-	0.18	0.10
Cu	-	-	9.48	3.41	-	-
Fe	9.15	1.78	0.24	0.09	0.50	0.39
Mg	-	-	-	-	3.13	2.53
Mn	-	-	-	-	0.52	0.35
Na	22.12	2.29	12.58	8.19	27.87	11.47
Ni	-	-	3.12	0.90	0.80	0.57
O	21.98	3.13	41.53	5.70	44.92	1.61
P	-	-	0.18	0.04	-	-
S	-	-	0.18	0.08	0.18	0.08
Si	-	-	0.18	0.04	0.25	0.17
Sn	-	-	24.92	7.77	-	-

D. Wastewater Analysis

Waste solutions were analysed using headspace SPME extraction coupled with GC-MS. Chromatograms of wastewater from 30 wt. % solutions are shown in Fig. 6. Dimethyl carbonate (peak A) and ethyl methyl carbonate (peak B) were the main studied compounds. Both were found in wastewater with 30 wt. % of NaCl but absent in wastewater with 30 wt. % of NaOH and $NaNO_3$. Sodium nitrate likely oxidized the carbonates, while sodium hydroxide hydrolysed it. Further investigation of degradation products and other compounds is suggested for future research.

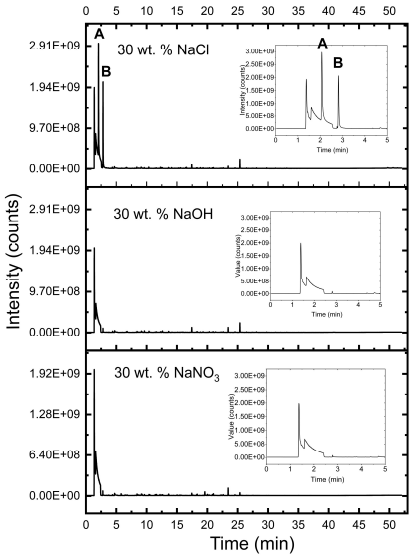

Fig. 6. Chromatograms of 30 wt. % of NaCl, NaOH, and $NaNO_3$ wastewater solutions.

E. Circular Economy Perspective

The study used results of released battery mass during electrochemical discharge to assess losses, focusing on Ni, Cu, Al, Fe, and Mn content. The released substance's minimum, maximum, and average quantities were determined based on Tab. II. Considering a typical cylindrical battery weight (50 g), the total released mass from 1 tonne of discharged cells was calculated. The estimated minimal and maximal levels of released monitored elements mass are shown in Tab. III.

TABLE III. ESTIMATION OF RELEASED BATTERY MASS AMOUNTS FROM 1 TONNE OF ELECTROCHEMICALLY DISCHARGED LIB CELLS.

Element	Minimal level (kg)	Maximal level (kg)
Ni	8.00	31.17
Cu	31.61	94.83
Al	20.33	55.67
Fe	2.40	91.50
Mn	1.72	5.17

The cost losses were determined using the current market price of the selected materials [6]; they cover the cost of the lost material if the waste solutions were disposed of as hazardous waste. A box chart illustrating the minimum, maximum, median, lower quartile, and upper quartile costs is provided in Fig. 7. In terms of economic losses, Cu accounts for the largest share, followed by Ni (released in approx. one-third of the amount compared to Cu). These results suggest the total cost losses range from 190 to 1560 USD per tonne of NCA LIB cells treated via electrochemical discharging.

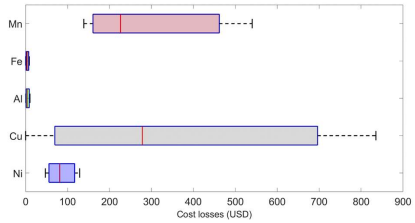

Fig. 7. Estimated cost losses due to released battery material during electrochemical discharging of NCA LIB cells.

IV. CONCLUSIONS

This study investigated the release of battery mass during the electrochemical discharge of LIB cells. Battery destruction and material release were observed, leading to potential cost losses of up to 1560 USD/tonne for NCA LIB cells. Moreover, electrochemical discharging has drawbacks like high toxicity, structure contamination, and material recovery complexities, necessitating caution before industrial implementation and promoting non-destructive discharge alternatives.

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6th International Conference on Nanomaterials Fundamentals and Applications
High Tatras, 16-19.10.2022

Lithium-Ion NMC Batteries: Chemical Toxicity Reflection of Wastewater and Scrap

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Abstract

The ever-increasing requirements for global carbon dioxide CO_2 emission reduction decrease the production of new internal combustion engine vehicles (ICEVs). On the contrary, the demand for electric vehicles (EVs) increases along with the number of spent lithium-ion batteries (LIBs). Recycling LIBs seems to be one of the most promising options for end-of-life (EOL) treatment solutions; however, many process effects of currently used battery compounds are still being addressed, e.g., the safety and potential risks of wastewater, battery scrap, or leaks into the air. In this work, the LIB nickel-manganese-cobalt (NMC) cell electrolyte was characterized in wastewater using gas chromatography with mass spectrometry (GC-MS), inductively coupled plasma with optical emission spectrometer (ICP-OES), and the residues of toxic substances bound to nm-µm valuable metal particles in battery scrap were determined by x-ray fluorescence (XRF).

Introduction

The ever-increasing requirements for global carbon dioxide CO_2 emission reduction resulted in decreased production of new internal combustion engine vehicles (ICEVs) and increased demand for electric vehicles (EVs) [1]. The EU responded to this fundamental change in the automotive industry by introducing new emission-free standards and regulations considering the waste treatment solutions, including the collection targets for portable waste batteries or recycling efficiencies for lithium-ion batteries (LIBs) reaching 65 % by 2025 and 70 % by 2030 [2].

The EVs' LIBs are typically based on valuable metals such as cobalt (Co), manganese (Mn), nickel (Ni), and lithium (Li) in the cathode, graphite (C) in the anode, and other essential materials such as aluminum (Al) and copper (Co) in packaging and electronic components. Lithium hexafluorophosphate ($LiPF_6$) based on toxic phosphorus (P) or fluorine (F) is commonly used as an electrolyte [3]. Further, the battery cell consists of organic polymers, binders, and other adhesives. Although recycling LIBs increases the raw materials saving and reduces the energy and environmental impacts of their mining, the effects of the entire process are still being addressed, such as the safety and potential risks [3,4].

The LIBs electrolyte consists of highly volatile and reactive compounds, resulting in highly toxic products, e.g., widely used $LiPF_6$ salt hydrolyses and evaporates solvents and HF. Many of these substances are released and taken away in wastewater; other residues then remain in a scrap of battery materials (pre-step of black mass production) and are released into the air [4].

This work is devoted to the first part of the characterization of LIB nickel-manganese-cobalt (NMC) cell electrolyte released in wastewater using gas chromatography with mass spectrometry (GC-MS). Major organic solvents were identified and relatively quantified by this technique. Inductively coupled plasma with an optical emission spectrometer (ICP-OES) was used for absolute quantification of selected elements eluted into water (Li, P, Ni, Mn, Co, Cu, Al). The residues of toxic substances such as P or F bound to nm-µm valuable metal particles in battery scrap using x-ray fluorescence (XRF). In addition, the residual proportion of substances after flushing was analyzed. According to the results, most of the electrolyte was released immediately upon breaking the cell cover; its share was already reduced to less than 10 % of total content during the first flush.

Methodology

This work used a deeply discharged NMC (712) pouch LIB cell with a nominal voltage of 3.65 V and 78 Ah capacity for laboratory treatment. This type of cell represents the building element of the Modular electric drive matrix (MEB) platform widely used in electric vehicles, e.g., Volkswagen, SEAT, Audi, or ŠKODA AUTO.

The proportion of valuable metals (Ni, Mn, and Co), covering materials (Cu and Al), and released cell's $LiPF_6$ electrolyte (monitoring by Li and P ratio) within the liquid medium (wastewater) was analyzed. Furthermore, the residual portion of toxic substances, focusing on P and F, in the waste scrap further processed into a black mass was determined. In addition, the reduction of these substances during repeated water-flushing was observed.

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Experiment procedure

After immersing the battery cell in a liquid (tap water), the Al cover was cut off (*cell opening*); individual layers were separated (*cell to layers*) and further crushed (*crushing*). After 30 minutes of rest at laboratory room temperature (23 °C), the solid phase was removed from the liquid (*solids removal*) and immersed in a new water bath. The flushing was repeated, overall, four times. The individual experiment steps are illustrated in Figure 1.

Chemical toxicity reflection

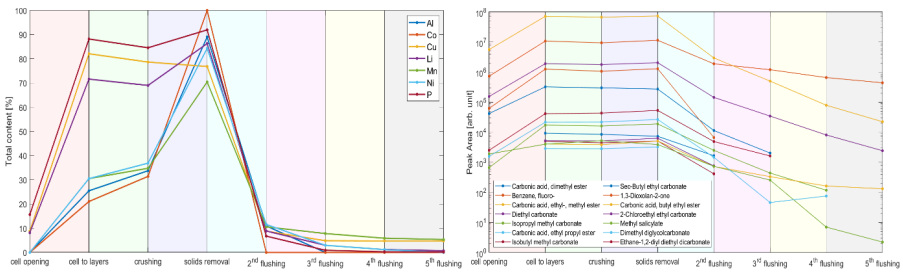
Gas chromatography with mass spectrometry (GC-MS) was utilized to identify major organic components released into wastewater. The applied method included Thermo Scientific Trace 1310 GC coupled to ISQ 7000 MS with the extraction of solvent from headspace over aqueous sample by acrylate SPME arrow performed by RSH autosampler. Incubation of samples was performed under 80 °C for 30 minutes. Extraction on the acrylate arrow ran for 5 minutes, thermal desorption in PTV inlet for 1 minute at 200 °C followed by chromatographic separation on TG-5SilMS column (30 m, 0,25 mm, 0,25 µm) with helium flow at 1,2 ml/min. The temperature program started at 40 °C and was held for 1 minute, followed by a linear temperature gradient of 5 °C/min up to 280 °C. The mass spectrometer was set to a 5 Hz scan rate with a range of 35-650 amu. To quantify selected elements released into the water, ICP-OES Perkin Elmer Optima 8000 was used. The samples were diluted 10x into 5% HNO3 and quantified towards 3-point calibration on Astasol multi-component standard. Samples over the linear range were further diluted in one order of magnitude. The nm-µm scale particles of material of thin layers on Cu and Al metal sheets were analyzed using the x-ray fluorescence (XRF) by the Philips PANAlytical spectrometer.

Results

The work analyzed the proportion of individual metals released into water, as shown in Figure 1 (a). When the cell was opened, the electrolyte was mainly released (characterized by the proportion of Li and P), its permissible exposure limit was set to 2.5 [mg/m^3] due to its high environmental and health risks as high acute toxicity, severe burns to skin and eyes. Repeated rinsing reduced its share to less than 1 % (0.7 for Li and 0.2 for P), as illustrated in Table 1. The metal release proportion depends on the individual layers' mechanical damage. At maximum comminution, the maximum release occurred. Flushing reduced the proportion of these materials in wastewater down to 5 %.

Metal emission into the air is expected to be negligible and is mainly present in the aqueous solution and solid parts. A low concentration of Co (less than 1 %) released into the water after the first flush shows its stability in the solid phase during the process; therefore, it possesses a minor risk for an aqueous environment. Cu is dissociating from the electrode into the solution at higher levels. Toxicity for higher mammals is low at these levels, but it may pose a threat to aquatic life. Background trace concentration of Co from tap water pipelines was observed. The detailed analysis of the obtained waste battery scrap characterizing the residues of P and F, as well as the valuable metals, will be covered in a full version of the manuscript.

Main hazards are induced by volatile organic solvents, especially fluorobenzene, esters of carbonic acid, and dioxolanone. They pose a threat due to high flammability, toxicity to aquatic life with long-lasting effects, eye/skin/respiratory irritation, or specific target organ toxicity. Long-term exposure to one of the main solvents, dimethyl ester of carbonic acid, was found to cause weight loss, altered sex ratio, increased number of stunted fetuses, and fetal malformations for rats at a level of 3000 ppm in air. Precautionary measures are therefore necessary for safe handling during recycling. An overview on these solvents is shown in Figure 1 (b). The substances were evaluated according to their hazard identification, including the GHS statements and LD50.



(a) (b)

Figure 1 Results: (a) wastewater analysis of metals in % of a total content, (b) volatile solvents released from wastewater.

Concentration (mg/l)	Al	Co	Cu	Li	Mn	Ni	P
cell opening	0.00	0.00	2.01	12.53	0.00	0.00	764.77
cell to layers	2.45	0.05	18.77	110.48	0.34	0.41	4316.84
crushing	3.25	0.08	17.99	106.44	0.39	0.50	4139.83
solids removal	8.58	0.25	17.57	133.13	0.79	1.15	4501.07
2. flushing	1.05	0.00	2.01	13.65	0.12	0.16	329.42
3. flushing	0.00	0.00	1.11	4.54	0.09	0.04	43.83
4. flushing	0.00	0.00	1.09	1.83	0.07	0.02	13.06
5. flushing	0.00	0.00	1.09	1.09	0.06	0.00	7.30
Sum	9.63	0.25	22.87	154.24	1.12	1.36	4894.68

Table 1 The concentration of target LIB cell elements released in wastewater.

Conclusion

Although recycling spent LIBs from EVs is considered one of the most promising EOL methods, the safety and potential hazards of wastewater, battery scrap, or leaks into the air are being necessarily addressed. Thus, this work characterized the release of a $LiPF_6$ electrolyte by determination of the lithium and phosphorus ratio in wastewater and battery scrap after detecting toxic phosphorus and fluorine. The results show that the electrolyte is dominantly released when opening the battery cell and it is almost fully taken away with water during the first flushing. According to the GC-MS, highly hazardous volatile organic solvents in the wastewater were detected; they were evaluated according to hazard identification, including the GHS statements and LD50. In future work, technological pathways for the purification of this water and material recovery will be proposed. This process's overall risks will be primarily characterized by respiratory exposure to dangerous substances. Based on our results, precautionary measures are necessary to implement for leaks into the air as well as proper liquid treatment and recycling of the solid metals.

Acknowledgements

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Unlocking Insights: A Systematic Survey of Material Composition in Lithium-Ion Battery Cells for Recycling Solutions

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***Abstract*—Battery recycling involves the recovery of materials from end-of-life (EOL) batteries, which are subsequently reused in the manufacturing of new products. Metals such as cobalt, lithium, nickel, manganese, aluminium, and copper are essential components of the electrodes of common lithium-ion batteries. In light of the growing demand for advancements in battery recycling, there is a critical need to start systematically documenting the chemistry of different types of batteries and to monitor the changes within their internal composition between the different states of charge, such as full charge, full discharge, deep discharge or shipping-state. In this work, a new methodology for a complex, quick, cheap, and effective material composition survey is presented. This methodology was applied to three types of cylindrical (18650) cells with two different cathode materials, specifically nickel cobalt aluminium (NCA) and nickel manganese cobalt (NMC), all with a capacity range from 3350 to 3500 mAh. This work offers a comprehensive, step-by-step description of the versatile battery research process, serving as the foundation for a streamlined and effective methodology for obtaining selected chemical and material parameters. The research endeavours to compare the material composition of lithium-ion cells at various states of charge to assess recycling potential and establish a database containing battery parameters. The results are essential for the automation and roboticization of the advanced recycling sector.**

Keywords—lithium-ion battery, end-of-life, recycling, sustainability, characterization, SEM-EDS

I. Introduction

Batteries provide portable power and energy storage options and are therefore indispensable in today's world. In particular, the use of lithium-ion batteries (LIBs) is growing across industries, putting pressure on recycling technologies [1, 2]. Thus, it is necessary to improve the recycling of valuable metals such as nickel, cobalt, manganese, lithium, aluminum, copper, and other minerals from batteries, from an economic and ecological point of view [3]. Several comprehensive works [4, 5] began with the study of the internal chemical composition and structures of selected types of commonly available batteries in different states, and special batteries from electric vehicles [6].

Nevertheless, it would be useful to develop an efficient, cheap, simple, and fast method for the comprehensive and safe analysis of batteries with different origins, chemical compositions and statuses, discharge levels or recycling potentials. If this method was sufficiently versatile, it could help to create an information database with monitored parameters, significantly speeding up the determination of recycling potentials. The comprehensive information collected in this way about batteries, considering their types or manufacturers, could speed up the development of advanced robotics and sophisticated automation of recycling lines and help optimize some recycling procedures, e.g., automatic and safe disassembly of battery packaging, monitoring of metal and mineral recovery, estimation of toxicity and losses leading to and decreasing the ecological impact, and increasing economic benefit.

II. Materials and Methods

A. Analysed battery cells

Three types of cylindrical (18650) LIB cells from the world's leading manufacturers, including SAMSUNG, PANASONIC, and LG, were selected to optimize a new methodology for fast, cheap, and efficient analysis of battery cells of different types and compositions. The cells were delivered in pristine condition and shipping-state of charge. Their characteristics are summarized in Table I. Prior to opening and analysis, the cells were not treated in any way, thus, the internal structure could not be affected. Two different types of chemical composition were examined: SAMSUNG and PANASONIC cells, with a general cathode composition of NCA (nickel cobalt aluminium), and an LG cell, based on the NMC (nickel manganese cobalt) spinel structure.

TABLE I. Characteristics of Selected Batteries

Battery cell	*Property*	*Tested state*	*Cathode chemistry*
SAMSUNG 35E	Capacity	3500 mAh	NCA
	Voltage	3.54 V	
PANASONIC NCR18650B	Capacity	3350 mAh	NCA
	Voltage	3.52 V	
LG DBM361865	Capacity	3440 mAh	NMC
	Voltage	3.53 V	

B. Methodology of samples preparation

The sample preparation process consisted of two consecutive parts. The first one consisted of the manual disassembly of the battery cells, where the safety was considered as the most critical aspect. During the disassembly, the cells were at original state of charge; nevertheless, after opening they self-discharged to the voltage below 1 V (deep discharge) within less than one day (24 hours). Deeply discharged battery cells can be physically cut off and the samples can be safely prepared.

The battery cell opening process involves making a straight cut to the cell casing around its circumference. Typically, a straight grinder with a ceramic cutting disc, which will prevent the battery from shorting and catching fire, is used. After completing the cut, the cells start to lose voltage; within 24 hours the voltage drops from around 3.5 V to about 0.5 V.

The samples preparation represents the second part of the procedure. A 5 mm high cylinder is cut from the split battery cell for a comprehensive cross-sectional chemical analysis of the entire battery. This cylindrical sample is manually ground on five different sandpapers (G120, G240, G400, G600, G1200) and dry-polished with a straight grinder polishing adapter. The first type of sample, a cross-section of the entire battery (SAMSUNG NCA LIB cell), illustrated in Fig. 1, is used to measure the thickness of the metal casing, the width of the individual layers of the anode and cathode, the separator, and to determine the complex chemical composition of the battery.

Fig. 1. A cross-sectional sample of the entire battery SAMSUNG NCA LIB cell, ready for measurement and chemical analysis.

The same part is cut off to measure the length of the whole electro-active strip rolled up in the battery. The last sample is cut, approximately from the middle of the cell, in the same way to prepare samples from the individual layers of the cathode and anode. The whole cca 5 mm wide strip of anode and cathode is unrolled and small pieces 5 x 5 mm from each electrode are cut from its centre. These pieces are taken two times from each layer, so each layer is then analysed from both sides. In the same way, the samples of the separator and especially the metal casing are prepared. These samples of cathode, anode, separator, and metal casing are carbon-taped to the holders, as shown in Fig. 2 (PANASONIC NCA LIB cell); they are also submitted for chemical analysis.

Fig. 2. Prepared samples of the cathode, anode, separator and metal casing from PANASONIC NCA LIB cell for chemical analysis (both sides).

C. Analysis

A Scanning Electron Microscope (SEM) TESCAN VEGA 3 LMU was used for image documentation, particle size and layer width measurement, and elemental composition determination, in secondary electron (SE) and backscattered electron (BSE) modes. The integrated VEGA3 software was used to measure the sizes, diameters of the particles, and the thickness of the layers. The OXFORD Instruments INCA 350 EDS analyser enables chemical microanalysis of the observed sample, which is subsequently evaluated in the AZtec software version 4.4. Using energy dispersive X-ray spectroscopy (EDS), the elemental composition was measured on an area of 0.005 mm^2 (magnification 2000x), 0.02 mm^2 (magnification 1000x), and 0.93 mm^2 (magnification 150x). Due to this, the relatively accurate chemical composition of the battery cells in their pristine state was effectively determined. The SEM-EDS settings for all measurements performed are summarized in Table II.

TABLE II. PARAMETERS OF SEM-EDS MEASUREMENT.

Parameter	*Values*
High Voltage	20 kV
Beam Intensity	15
Working Distance	15 mm
Magnification	150x, 1kx, 2kx
Vacuum in Chamber	9.4-8.7 $x10^{-2}$ Pa
Vacuum in Tubus	5.8-5.6 $x10^{-2}$ Pa
Detector	SE, BSE

A list of battery parts, along with the numbers of measurements, provided in Table III, was created, and studied on each battery, which would be necessary to optimize the rapid analysis process for recycling purposes.

TABLE III. TABLE OF NUMBERS OF MEASUREMENTS.

Properties	Table of Measurements	
	Battery parts	*Number of measurements*
Proportions	Inner electrode strip	2
Thickness	Metal casing	10
	Cathode	20
	Active layer (NCA, NMC)	20
	Al collector	20
	Anode	20
	Active layer (C, C+Si)	20
	Cu collector	20
	Separator	20
Composition	Metal casing	4
	Cross-section	6
	Active layer (NCA, NMC) A side	6
	Active layer (NCA, NMC) B side	6
	Active layer (C, C+Si) A side	6
	Active layer (C, C+Si) B side	6
	Separator	4

The length and width of the electroactive strip inside the battery were measured manually with a ruler. Using image analysis of the sample of the first type (cross-section), the thickness of the metal casing and the width of the inner layers were studied at a magnification of 100x, resp. 150x. The results were obtained using a software tool, and each layer was measured at least twenty times. Using SEM-EDS in high vacuum, the average composition of the battery interior was also determined on cross-sectional samples. This composition was measured six times from an area of 0.93 mm^2. The work results represent the average values of measurements.

The chemical composition of the individual active layers was measured from both sides of the electrode (side A and side B) at a magnification of 1kx (a total of three measurements on each side) and at a magnification of 2kx (also a total of three measurements on each side). Since both sides of one electrode should have the same composition, the average value is calculated from a total of twelve measurements. The magnification used for image analysis and the number of chemical composition measurements are shown in Table IV. The metal casing mostly had a simple composition which is the same on both sides, therefore only four measures at two different magnifications were sufficient. In the case of visible corrosion products from the inside of the battery, the composition was analysed outside of these products. The corrosion products will be studied in more detail in the following work.

TABLE IV. MAGNIFICATION OF IMAGE AND COMPOSITION ANALYSIS.

Properties	**Table of Magnifications**	
	Battery Parts	***Magnification (number of measurements)***
Proportions	Inner electrode strip	0
Thickness	Metal casing	100x
	Cathode	150x, 600x
	Active layer (NCA, NMC)	150x, 600x
	Al collector	150x, 600x
	Anode	150x, 600x
	Active layer (C, C+Si)	150x, 600x
	Cu collector	150x, 600x
	Separator	150x, 600x
Composition	Metal casing	1kx (2x), 2kx(2x)
	Cross-section	150x (6x)
	Active layer (NCA, NMC) A side	1kx (3x), 2kx(3x)
	Active layer (NCA, NMC) B side	1kx (3x), 2kx(3x)
	Active layer (C, C+Si) A side	1kx (3x), 2kx(3x)
	Active layer (C, C+Si) B side	1kx (3x), 2kx(3x)
	Separator	1kx (2x), 2kx(2x)

The same procedure was applied to a polymer separator that could not be identified specifically by EDS. This measurement is mainly supplementary and helps to distinguish pure polymer from polymer with a surface treatment (e.g., aluminum oxide).

III. RESULTS AND DISCUSSION

This section is divided into two parts, which successively present the results obtained by the optimized methodology of fast battery survey using SEM-EDS. In the first part, there are the results of measuring the proportions and thickness of the electroactive layers, in the second part, the results of the chemical composition are presented and their reliability and time-consuming are described. Together with the results, the individual steps of the optimized methodology are continuously discussed, and their effectiveness is evaluated.

A. Proportions and thickness of layers

Before the analysis itself, all available parameters were measured (voltage and size of the battery, which coincides with the width of the internal electroactive strip without metal casing). Subsequently, the metal casing of each battery was opened as described in the previous section. From the discharged cells, the samples for chemical composition analysis were prepared. The measured values are shown in Table V, where the full length of the electroactive strip is presented via its entire area (length x width) in mm^2. The values in Table 5 are averaged from more than 20 measurements and, for practical reasons, rounded to the nearest whole number within the standard deviation for further calculations.

TABLE V. THICKNESS, WIDTH, AND AREA OF TESTED CELLS.

	SAMSUNG 35E	***PANASONIC NCR18650B***	***LG DBM361865***
Cathode (μm)	150	180	150
Cath. active layer (μm)	60	75	60
Al - collector (μm)	30	30	30
Anode (μm)	140	180	150
Anod. active layer (μm)	50	75	55
Cu – collector (μm)	40	30	40
Separator (μm)	20	20	20
Metal casing (μm)	200	150	260
Electroactive strip, area (mm^2)	700x63	680x63	660x63

The surface area of the entire internal electroactive part of the battery, which indicates the value of the capacity and therefore the quality of the battery, is in all cases around 400 cm^2. The thickness of the metal casing, which also serves as the negative electrode, was measured using the integrated software in the VEGA program (Fig. 3).

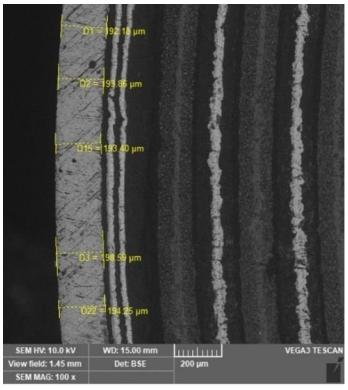

Fig. 3. An example of measuring metal casing of SAMSUNG NCA LIB cell (magnification 100x) on a battery cross-section.

The layers of the cathode, anode, and separator in cross-section were measured in the same way (Fig. 4). A quick determination of the thickness of the metal casing, or even a database of the most used types of metals and metal casings, can be very useful for automating the disassembly process of batteries with unknown conditions. The thickness measurement was performed several times due to the large standard deviation. When preparing the sample for this measurement, the quality of the grinding and polishing of the sample according to metallographic procedures is very important, in this way the standard deviation can be greatly reduced. After high-quality sample preparation, it is enough to average 20 values, which are summarized in Table V.

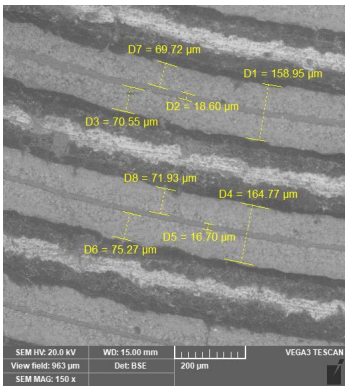

Fig. 4. Illustration of the thickness material layers measurement for cathode layers, aluminum current collectors, and separators of the SAMSUNG NCA LIB cell (magnification 150x).

B. *Material composition analysis*

The composition was measured on a cross-section, for a quick comprehensive analysis (Table VI), and on the surface of individual layers to accurately determine the mass of individual metals, the stoichiometry of assumed minerals, and the relative representation of other elements on the surface of cathode (Table VII) and anode (Table VIII).

TABLE VI. AVERAGE COMPOSITION ON CROSS-SECTION.

Elements (wt. %)	***SAMSUNG 35E***	***PANASONIC NCR18650B***	***LG DBM361865***
C	51.02±0.54	51.90±2.73	47.10±2.25
O	12.40±0.35	14.51±1.51	17.78±0.74
Cu	10.08±0.48	8.11±2.73	6.90±0.88
Ni	10.52±0.40	11.29±2.11	11.7±1.43
Al	7.96±0.70	3.56±1.07	5.03±0.29
F	4.08±0.29	5.36±1.21	6.33±2.13
Co	1.42±0.04	2.32±0.48	1.50±0.16
Fe	0.48±0.04	1.55±0.82	0.50±0.34
P	0.50±0.02	1.54±0.46	0.58±0.10
Si	1.10±0.14	0.18±0.04	0.78±0.05
S	0.02±0.04	0.17±0.05	1.00±0.22
I	0.34±0.05	0.10±0.26	
Mn			0.83±0.13

The EDS mapping of the cross-section of the battery at 150x magnification is presented in Fig. 5. Fig. 6 and Fig. 7 show the active layers of the cathode and anode at the highest used magnification (2kx).

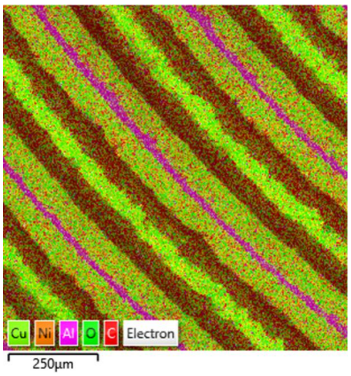

Fig. 5. Example of The EDS mapping of the average elemental composition inside the SAMSUNG NCA cell, measured over an area of 0.93 mm^2.

Not only are the thicknesses of the individual layers determined on the cross-section of the battery, which together with the area corresponds to the volume of the chemical components, but the specific average weight of the composition inside the battery is quickly determined in this way. Mainly pure metals from current collectors (Cu, Al) and valuable metals from the cathode layer are monitored (specifically the content of Co, Ni, Mn). The composition of the samples needs to be measured in a high vacuum in order to be as accurate as possible. Repeated measurements were able to reproduce even very complex compositions with a very small deviation. These results can be further used to calculate the recycling potential of individual batteries, etc.

TABLE VII. COMPOSITION OF CATHODES.

Cathode (wt. %)	*SAMSUNG 35E*	*PANASONIC NCR18650B*	*LG DBM361865*
Co	5.45±0.10	9.68±0.45	6.52±0.25
Ni	33.73±0.54	50.11±1.22	47.68±2.02
Al	1.50±0.13	0.98±0.11	0.23±0.1
Mn			3.43±0.08
O	22.73±0.51	26.62±1.78	27.82±1.85
C	23.78±0.90	10.43±1.13	9.82±0.52
F	12.13±0.81	3.82±0.40	4.04±0.52
P	0.35±0.06	0.31±0.19	0.18±0.08
S	0.10±0.01		0.40±0.06

The composition of the electroactive layers was performed from both sides of each electrode to exclude material gradients or unexpected changes. However, the results showed that for all types of batteries, both sides of the cathode and anode logically are completely the same. A difference of this type was noted only in the case of the separator, it is due to the one-sided modification of the polymer for electrical insulation.

The main monitored element on the cathodes of all types of batteries is cobalt (expensive and toxic). The amount of cobalt is a suitable indicator for economic and ecological calculations. The results showed that all batteries contain a high percentage of cobalt, and in the case of the LG battery, cobalt-enriched spinel NMC 712 is even used. In both the NCA and NMC types, nickel makes up a relative half of the weight. It is noteworthy that identical levels of fluorine were detected across all cases, likely originating from both the polymer binder within the active layer and residual electrolytes.

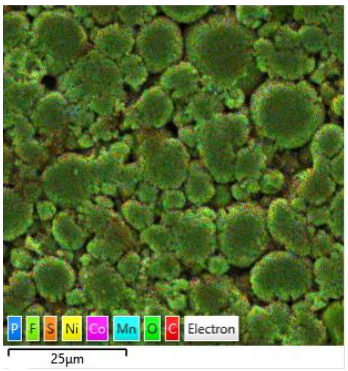

Fig. 6. Example of the EDS mapping of the average elemental composition of cathode inside the LG NMC LIB cell, measured over an area of 0.005 mm^2.

The surface of all the anodes is compared from the point of view of the content of silicon particles pressed into the graphite to increase the battery capacity without showing defects associated with the volumetric expansion of silicon. The content of silicon particles in graphite can be an indicator of battery quality, and at the same time, this information is useful for recycling and material separation needs.

TABLE VIII. COMPOSITION OF ANODES.

Anode (wt. %)	*SAMSUNG 35E*	*PANASONIC NCR18650B*	*LG DBM361865*
C	90.49±0.64	99.25±0.12	86.68±0.76
P	0.31±0.02	0.63±0.07	0.28±0.05
Cu	0.76±0.05	0.13±0.08	0.70±0.02
Si	2.49±0.64		2.58±0.29
F	1.33±0.10		1.43±0.19
O	4.59±0.29		7.68±0.46
S			0.68±0.05

The EDS method is very suitable for fast and accurate composition research, here it has been proven that the results of measuring the composition of properly prepared samples are the same at any magnification used (here 1kx or 2kx). From a methodological point of view, however, it is necessary to observe the measurement while gradually increasing the magnification, because with a more complex composition or with samples that are not in a pristine state, even minor components can be detected at a higher magnification. In this simple way, it was found that LG (NMC) and SAMSUNG (NCA) batteries use the same type of anode enriched with silicon particles that increase the capacity of the entire layer. PANASONIC NCR18650B, on the other hand, still uses pure graphite technology. The results are again the average from the measurements of both sides of the anode layer. The same composition was confirmed on both sides as expected.

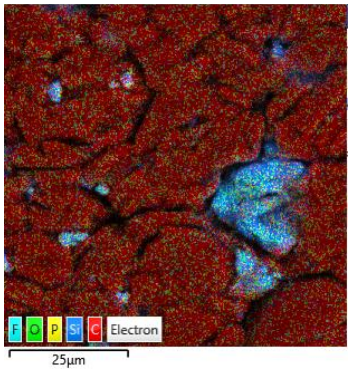

Fig. 7. Example of The EDS mapping of the average elemental composition of anode inside the SAMSUNG NCA LIB cell, measured over an area of 0.005 mm^2.

In the same way and under the same SEM-EDS setup conditions, the composition of the other parts of the battery was also determined: the metal casing and the separator. The metal casing was identified in all cases as an alloy of nickel and iron with a mixture of carbon (Table IX). They differ from each other only in their thickness.

TABLE IX. COMPOSITION OF METAL CASINGS.

Elements (wt. %)	***SAMSUNG 35E***	***PANASONIC NCR18650B***	***LG DBM361865***
Ni	88.15±0.21	87.23±0.26	84.05±1.06
Fe	5.95±0.26	6.68±0.24	6.25±0.49
C	5.75±0.07	6.08±0.38	6.10±0.28
O			3.50±0.57
Al	0.10±0.02		

In the case of the separator, the measurement corresponded to the assumption that one side of the separator is modified with aluminium oxide for practical reasons, Table X, and Table XI shows the results of the elemental composition from both sides separately.

TABLE X. COMPOSITION OF SEPARATORS (MODIFIED SIDE).

Elements (wt. %)	***SAMSUNG 35E***	***PANASONIC NCR18650B***	***LG DBM361865***
Al	50.20±5.94	20.45±0.35	31.95±5.59
O	42.65±3.18	29.90±0.90	32.65±7.14
F	2.50±0.10	11.50±0.14	26.50±12.02
C	10.40±0.40	34.10±0.14	6.90±0.28
S			1.10±0.14
P	0.65±0.07	3.50±0.02	0.90±0.28
Na		0.50±0.01	

TABLE XI. COMPOSITION OF SEPARATORS (NON-MODIFIED SIDE).

Elements (wt. %)	***SAMSUNG 35E***	***PANASONIC NCR18650B***	***LG DBM361865***
C	90.40±0.05	94.55±0.35	86.30±0.85
F	6.95±0.49	4.20±0.28	6.00±0.28
O			4.00±0.28
S			1.75±0.21
P	1.40±0.02	1.05±0.07	1.35±0.07
Al	1.15±0.49	0.05±0.03	0.30±0.02
Na	0.20±0.01	0.10±0.01	0.10±0.01

IV. CONCLUSION

Using the presented methodology, three types of commercial cylindrical battery cells with a shipped-state voltage level of 3.5 V were chemically and materially analysed. Two types of cells in NCA and NMC composition were selected for mutual comparison. The presented methodology has still a space for improvement, but the current state already shows results with a very high potential for wider application in industry with universal use. This approach is primarily characterized by safe and relatively quick obtaining of the necessary results. The presented measurement and elemental analysis of the internal structure of battery cells based on the SEM-EDS method is reliable, fast, reproducible, and with a very small deviation if the recommended settings are followed. The presented step-by-step procedure aspires to a generalized methodology for a fast and effective research of the recycling potential of battery cells and for the creation of a database of technical and chemical parameters of all available batteries for the needs of advanced automation and robotization of recycling lines. The results from this work will be further applied to the calculations of the recycling potential and subsequently compared with the actual yield. This work represents a baseline for the following research, including a detailed description of the self-discharging process during disassembly, determinations of the content of individual elements, and evaluation of their economic benefit, ecological impact, and inner corrosion of metal casing.

ACKNOWLEDGMENT

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Lithium-ion battery module-to-cell: disassembly and material analysis

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Abstract. Lithium-ion batteries (LIBs) are one of the most popular energy storage systems. Due to their excellent performance, they are widely used in portable consumer electronics and electric vehicles (EVs). The ever-increasing requirements for global carbon dioxide CO_2 emission reduction inhibit the production of new combustion vehicles. Thus, the demand for EVs increases, as well as the number of spent LIBs. Due to increases in raw materials saving and reduction in energy and environmental impacts, recycling is one of the most promising solutions for end-of-life (EOL) treatment for spent LIBs. This work describes the first step in recycling the LIBs nickel-manganese-cobalt (NMC) based module from a full battery electric vehicle (BEV) holding its high recycling efficiency and considering the process costs and environmental impact. This paper is devoted to module-to-cell disassembly, discharge state characterization measurements, and material analysis of its components based on x-ray fluorescence (XRF) and diffraction (XRD).

1. Introduction

Lithium-ion batteries (LIBs) currently represent one of the most promising electrochemical energy storage systems. They are widely used in a broad range of applications, from powering small consumer electric devices such as smartphones or laptops to integrating into electric transport systems considering electric bikes or scooters and electric vehicles (EVs). Moreover, LIBs are one of the key technologies for stationary energy storage systems [1,2].

The automotive industry is undergoing a fundamental change. The ever-increasing requirements for global carbon dioxide CO_2 emission reduction led to a decrease in the production of new combustion vehicles and an increase in the demand for electric ones [3]. In Europe, the whole emission-free concept shield within the European Green Deal is supported by Regulation (EU) 2019/63; it sets EU fleet-wide emission performance standards for new passenger cars and new light commercial vehicles (vans) registered in the EU. Moreover, the Fit for 55 legislative package presents EU's ambitious target of achieving net emission reductions of at least 55% by 2030, compared to 1990 levels [4].

The electric vehicle LIBs are generally based on valuable metals such as cobalt (Co), manganese (Mn), nickel (Ni), and lithium (Li), other metals such as iron (Fe) and aluminium (Al) or the non-metallic elements such as phosphorous (P) in the cathode, other elements such as graphite (C) in the anode, and materials such as Al, copper (Cu), and Fe in coverings and electronic components. The electrolyte of

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LIBs is typically made up of lithium salt solution such as lithium hexafluorophosphate $LiPF_6$ salt. Additionally, the battery consists of organic polymers, e.g., a separator, binders, and adhesives [5,6].

The constant growth in the EVs market leads to the ever-increasing number of spent LIBs and brings numerous waste-management challenges in end-of-life (EOL) processing. Due to increased requirements on raw materials savings and reduction in energy and environmental impacts of mining, recycling is one of the most promising solutions for battery waste treatment [7]. A proper recycling technique recovers selected materials with high efficiency and purity, reaching more than 90 % [8]. The EU responded to the current situation and issued a proposal for a Regulation of the European Parliament and the Council concerning batteries and waste batteries, repealing Directive 2006/66/EC and amending Regulation (EU) No 2019/1020. The proposal has established, e.g., collection targets for portable waste batteries, which are 70 % by the end of 2025, instead of the current 65 %, and 80 % by the end of 2030; it regards recycling efficiencies for LIBs treatment: 65 % by 2025, 70 % by 2030 [9].

This work represents the first step in recycling the LIB module while maintaining its high efficiency and considering the process's overall cost and environmental impact. It focuses on the disassembly of the LIB module typically used in a full battery electric vehicle (BEV) to its NMC-based cells and provides the material analysis. Electrochemical impedance spectroscopy (EIS) and five cycles of cyclic voltammetry (CV) were measured in terms of safety and complex discharge state characterization of the cell. The detailed analysis of the material composition of the battery cell components was determined using x-ray fluorescence (XRF) and diffractometry (XRD). According to the analysis, the content of valuable metals in the N:M:C ratio was determined as 54%:23%:23%. The difference between the proportions of these metals could affect the recycling process's overall economic and environmental impact and will be studied in more detail in future work.

2. Experimental

This paper presents performed dismantling treatment and the material analysis on the battery module's scale to its cells. The LIB module typically used in the battery electric vehicle (BEV) was chosen for laboratory testing. The module consists of 24 NMC pouch cells in a 3P8S connection, each with 3.65 V and 78 Ah capacity, which create the base for the nominal module voltage of 29.36 V.

2.1. Disassembly procedure

For an evaluation of the process complexity, the module was manually disassembled into individual parts. Before disassembling, the module was fully discharged and short-circuited to ensure safe handling. Firstly, the Al packaging was milled off. It was followed by removing the insulating and thermo-insulating materials, which were later analyzed as commonly used polyurethane and polymer based on a mixture of aryl phosphates and hydrated aluminum oxides, with the presence of rutile (TiO_2) and corundum (Al_2O_3). Secondly, the remaining plastics and metal coverings were manually split off, and finally, the individual cells were separated. The treated module is shown in Figure 1 (a), the removed covering materials are illustrated in Figure 1 (b), and the separated battery cell is in Figure 1 (c).

2.2. Battery cell testing

For a characterization of the discharged state of the cell after short-circuiting the entire module, one of the separated battery cells has undertaken a series of measurements, including the EIS and CV. These measurements are used to describe the migration of charge carriers, characterize the interfacial properties of materials at the electrode surface and identify the electrochemical reactions of active electrode materials.

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(a) (b) (c)

Figure 1. Example of (a) treated BEV module, (b) removed coverings, and (c) battery cell.

2.2.1. Electrochemical impedance measurement

The EIS measurement is commonly used for investigating the properties of materials or electrode reactions by measuring the impedance over a range of frequencies. The complex impedance Z^* was recorded with an Autolab PGSTAT30 potentiostat using a harmonic AC field from 1 MHz to 1 mHz at an amplitude of 1 mV. The absolute value of impedance /Z/ and its phase angle φ were plotted in a Bode plot as functions of frequency. For the spectra analysis, the negative imaginary part $-Z_{Im}$ was plotted vs. the real part Z_{Re}, thereby forming a Nyquist plot on which frequency is a function parameter.

An equivalent circuit model (ECM) with electric components whose parameters quantify the physicochemical processes inside the cell under the electric field can be extracted from an impedance spectrum obtained via EIS. Based on the plots shape, a fractional-order ECM for LIB, which is shown in Figure 2, was used [10]. Using the non-least-squares method (NLSM), the impedance data were fitted to the equivalent circuit and the parameters of circuit components were calculated.

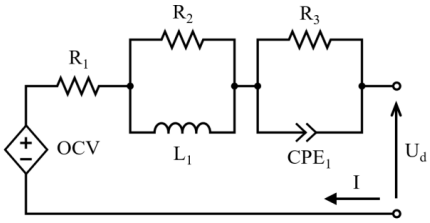

Figure 2. A fractional-order equivalent circuit model for LIB cell.

In the circuit, the resistor R_1 represents the high-frequency resistance of the electrolyte $LiPF_6$ and its impedance Z_1 was expressed as:

$$Z_1 = R_1. \tag{1}$$

Because the battery cell was in a completely discharged state after it had been short-circuited, the value of the original electrolyte resistance might be additionally lowered by the post formation of highly conductive “channels” during this discharge process.

The parallel combination of the resistor R_2 and inductor L_1 represents high-frequency inductive behavior and its impedance Z_2 was determined according to equation (2).

$$Z_2 = \left(\frac{1}{R_2} + \frac{1}{i\omega L_1}\right)^{-1} \tag{2}$$

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The inductive response is probably caused by the layered structure of metallic foils inside a battery cell or by the prevailing electronic conductivity of cathode or anode materials.

The parallel combination of the resistor R_3 and constant phase element (CPE) represents the distributed capacitance response to the migration of charge Li^+ carriers and electrochemical reactions in electrode materials. Its impedance Z_3 was calculated as:

$$Z_3 = \left[\frac{1}{R_3} + A(i\omega)^{\alpha}\right]^{-1}, \tag{3}$$

where A is the distributed capacity in an electrode and at the electrode/metal collector surface, and α is a parameter characterizing the distribution. The total impedance of the cell Z was calculated as a sum of the partial impedances above:

$$Z = Z_1 + Z_2 + Z_3. \tag{4}$$

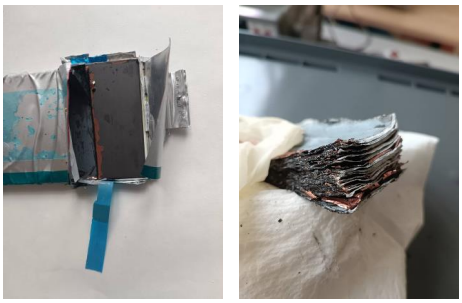
2.2.2. Cyclic voltammetry measurement

The CV is typically used for the analysis of electrochemical redox reactions of active materials by measuring the current as a function of potential. In this work, this was also recorded with an Autolab PGSTAT30 potentiostat in the staircase mode from 0 V to the potential when the current reached the value of 1 A; the potential step was 2.44 mV with scan rate 2 mV/s.

The plots of current I vs. potential E indicate the change of polarization resistance R_p during the potential cycles when a total of five cycles were measured. The derivative of recorded CV curves dI/dE represents the reciprocal polarization resistance $1/R_p$ of the electrochemical reactions inside a battery cell. Moreover, the hysteresis of the curves indicates the difference of R_p due to the charging or discharging process. Such CV measurements are very useful as an EIS complement for precise determining the polarization resistance R_p which could relatively indicate the state of health (SOH) of a spent battery cell.

2.3. Material analysis

For further processing, the battery cell was opened and dismantled to its individual layers of electrode materials soaked with electrolyte. The material of these thin layers on Cu, and Al metal sheets or separator were analyzed using the x-ray fluorescence (XRF) using the Philips PANAlytical spectrometer and x-ray diffraction (XRD) using Philips PANAlytical XPERT-PRO diffractometer. The composition of cathode, anode and electrolyte materials were determined, and the material ratio of valuable metals (Ni:Mn:Co) was discussed. The first cut in a battery cell is shown in Figure 3 (a); the individual layers of the cell's electrode materials are demonstrated in Figure 3 (b).



(a) (b)

Figure 3. Example of (a) a cut in battery cell, (b) individual layers of electrode materials.

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3. Results and Discussion

The module was disassembled manually using a milling cutter, pliers, and other specialized tools. Due to this was the first processing of this technology, the process was complicated, and the total disassembly time was around 3 hours. The separation of one battery cell, including adhesive cleaning with ethanol, was about 30 minutes on average.

The discharge state of the cell was evaluated using EIS measurements. In the frequency range of 2 kHz to 10 Hz, the impedance exhibited ohmic character with φ close to zero. For the frequencies above 2 kHz, the character was inductive ($\varphi > 0$), and below 10 Hz the character was capacitive ($\varphi < 0$). The distribution of capacity A is characterized by the parameter $0.5 < \alpha < 1$ which indicated the formation of a space charge in the region close to the metallic current collectors of a battery cell. This space charge was disturbed by the migration of charge carriers; when $\alpha = 1$ than the electrochemical system behaves as an ideal capacitor, when $\alpha = 0.5$ than the system response is mainly controlled by the diffusion of ions. The measurements are shown in Nyquist and Bode plots illustrated in Figure 4. The relatively low resistance values for resistors R_1, R_2 and R_3 indicate a high electric conductivity of the electrolyte and electrode materials that form thin layers of the cell. The values could be also affected by the deep discharge process.

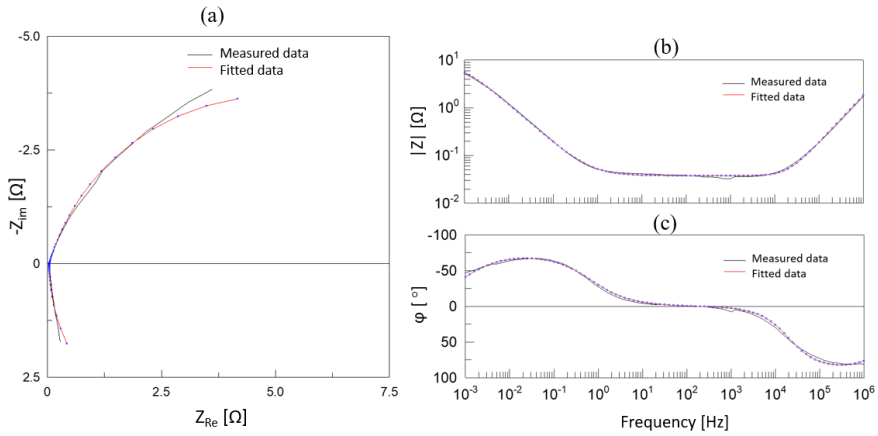

Figure 4. Impedance measurement (a) Nyquist impedance plot; Bode plots of (b) absolute value of impedance /Z/, and (c) its phase angle φ as functions of frequency.

The parameters of the ECM were calculated and are given in Table 1.

Table 1. Established ECM parameters according to the NLSM fit.

Parameter	Fitted value	Error	Relative Error [%]
R_1 [mΩ]	37.68	0.27	0.72
R_2 [mΩ]	8.34	1.06	12.72
R_3 [mΩ]	9.78	0.46	4.69
L_1 [μH]	0.29	0.33	1.12
A [F]	8.05	0.09	1.15
α [-]	0.82	0.01	0.55

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The resistance calculated from CV data dynamically changed during the potential cycle in the range of 10 Ω to 1 Ω. The lowest resistance values were achieved for the highest current values at the high and low vertex potentials, which is demonstrated in Figure 5. The observed extent of hysteresis indicates the significantly decreased reversibility and SOH of the spent battery cell in the measured potential window.

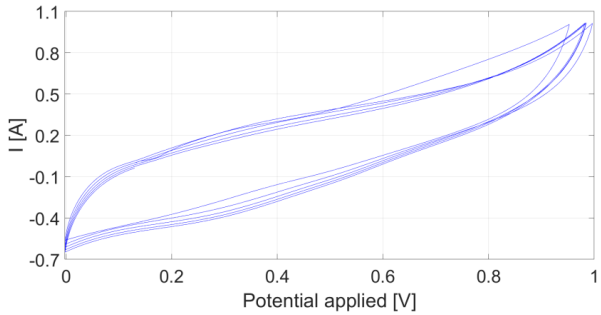

Figure 5. CV plots for five cycles.

A detailed characterization of the cell composition is an initial step to determine the suitable recycling procedure. A proportion overview of individual materials, especially metals, is necessary to evaluate the effectiveness of the selected technique or to define the expected economic return. In this work, the x-ray fluorescence (XRF) and x-ray diffractometry (XRD) analysis for the material composition determination was used; its results are summarized in Table 2.

Table 2. Determined material composition of the battery cell.

Component	Material composition
Cathode	Al sheet, $LiMn_{0.15}Co_{0.15}Ni_{0.7}O_2$
Anode	Cu sheet, C (min. 90 %) mixed with $LiCo_{0.25}Ni_{0.75}O_2$ and CuO or Al_2O_3
Electrolyte	$LiPF_6$
Separator	polymeric organic thin film

Cathode founded on Al sheet is created by mixed oxide of stoichiometry $LiMn_{0.15}Co_{0.15}Ni_{0.7}O_2$ and contains an amorphous phase, which is probably a binder of organic composition. Other impurities can be due to the dissolution of the surroundings and transport by diffusion into the cathode mass. These impurities are low in the amorphous phase. Anode based on Cu sheet consists of more than 90 % graphite with a 1 to 3 wt% admixture of lithium mixed oxide of stoichiometry $LiCo_{0.25}Ni_{0.75}O_2$ and copper oxide CuO and possibly aluminum oxide Al_2O_3. These impurities are probably caused by the partial dissolution of the surrounding components in the electrolyte and their subsequent diffusion into the electrode mass. The electrolyte represents a commonly used lithium hexafluorophosphate $LiPF_6$. The separator is created by a polymeric organic thin film, which has not yet been analyzed in detail. Moreover, the ratio between Ni, Mn, and Co (N:M:C) in the cathode material was determined and was closest to the values of 54%:23%:23%. It can be assumed that this proportion between the main elements is used to achieve the most suitable properties of the battery cell.

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4. Conclusions

Recycling is one of the most promising disposal methods for spent LIBs, especially those from EVs, which contain a considerable proportion of valuable metals such as Co, Ni, and Mn. Therefore, it is necessary to optimize current recycling techniques to achieve the highest possible proportion of recovered materials of sufficient quality for further use.

This paper represents the first step in the recycling process of a LIB module based on NMC cells. Short-circuiting the entire module was identified as a sufficient way to discharge individual cells to their safe limit, which is suitable for further handling. The material analysis results determined the proportions of metals and other elements of the battery cell compounds, which are necessary for further processing, including crushing, particle separation, and final recovery of valuable metals. The ratio between Ni, Mn, and Co (N:M:C) in the active cathode material was established and was closest to the values of 54%:23%:23%. In future work, a more detailed analysis of more battery cells will be provided. The results will be included in the final environmental and economic evaluation of the BEV modules, considering the production and waste treatment.

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Ultrasonic-Assisted Electrode Material Separation: Towards Industrial Applications

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Abstract*—The efficient recovery of valuable materials from spent lithium-ion batteries (LIBs) is essential for sustainable recycling and resource conservation. Ultrasonic-assisted separation has emerged as a promising recycling pre-treatment technique, which offers significant potential to enhance the material recovery processes. Thus, this study investigates the effectiveness of ultrasonic-assisted separation of electrode materials from lithium nickel manganese cobalt oxide (NMC 622) LIB pouch cells and evaluates the feasibility of scaling this method for industrial applications. Among the solvents tested (ethanol, acetone, isopropyl alcohol, N-Methyl-2-pyrrolidone (NMP), and distilled water), distilled water demonstrated the highest separation efficiency, achieving 69.4% removal of cathode material and 72.6% removal of anode material. Organic solvents like ethanol and acetone showed moderate performance. In addition to laboratory-scale efficiency, this research addresses critical challenges related to industrial implementation, such as limitations in sample loading, solvent accessibility, and process scalability. The number of samples processed in each batch significantly reduces separation efficiency due to material overlap and limited solvent interaction. This indicates the need for process modifications, such as improved agitation or optimized flow control. Furthermore, while ultrasonic separation presents an environmentally friendly alternative to chemical-intensive methods, ensuring its robustness for large-scale operations requires further optimization. This study offers valuable insights into both the fundamental performance of ultrasonic-assisted separation and the practical barriers to its industrial adoption, contributing to the advancement of more efficient and scalable LIB recycling technologies.

***Keywords*— material separation, lithium-ion battery, recycling pre-treatment, industrial applications**

I. Introduction

The efficient recovery of materials from spent lithium-ion batteries (LIBs) is critical for advancing sustainable recycling and resource conservation, particularly as global demand surges due to the rise of electric vehicles, portable electronics, and renewable energy systems [1], [2]. Effective recycling processes are structured into multiple stages, with the pre-treatment phase playing a pivotal role [3], [4]. This phase involves a combination of mechanical and chemical operations designed to prepare battery components for subsequent hydrometallurgical or pyrometallurgical extraction methods [3]. By optimizing processes such as disassembly, discharging, crushing, and separation, pre-treatment improves recovery efficiency and material purity and addresses environmental concerns and operational costs associated with recycling [3], [5]. Among innovative pre-treatment techniques, ultrasonic-assisted separation has emerged as a promising technology due to its potential to improve the efficiency and selectivity of active materials from the surfaces of current collectors [6].

The ultrasonic-assisted separation technique uses high-frequency sound waves to create cavitation bubbles in a liquid medium. When these bubbles implode, they generate localized high temperatures and pressures, which produce strong micro-jets and shock waves that enhance the separation process [6]. Ultrasonic treatment has several advantages over traditional mechanical and chemical methods, including reduced energy consumption, minimal chemical usage, and the ability to process delicate materials without significant structural damage [5].

Several studies have demonstrated the effectiveness of ultrasonic techniques in recovering active materials from LIB electrodes, typically within the laboratory-scaled application. A peeling-off ratio of up to 91.34% was achieved by applying ultrasonic treatment at a frequency of 40 kHz, combined with mechanical stirring, to 1 g of cut copper negative electrodes from 18650 cylindrical cells dispersed in 200 ml of water [6]. In study [7], 2 Ah pouch cells with lithium nickel manganese cobalt oxide (NMC 622) cathodes and graphite anodes were treated. Using a sequential separation technique with dimethyl carbonate, KH_2PO_4 buffer, and Triton™ X-100 surfactant, the separation efficiency for electrode materials was nearly 100%. Another work [8] also investigated the ultrasonic delamination of electrode materials containing NMC and lithium manganese oxide (LMO) using a high-power ultrasonic unit. The process achieved a material recovery rate of 99.5% for both electrodes, with only 0.5% loss attributed to small carbon black particles.

Despite recent advances in battery recycling, existing research has primarily focused on laboratory-scale applications [9] and has not comprehensively evaluated the scalability of processes, solvent efficiency, and material recovery limitations in industrial settings. Furthermore, current findings do not indicate a clear trend in optimal separation conditions, as different studies have utilized various approaches depending on the electrode material and binder composition. To address these inconsistencies, this study investigates the ultrasonic-assisted separation of electrode materials from NMC 622 LIB pouch cells, emphasising industrial feasibility. A selection of standard chemical solvents, along with N-methyl-2-pyrrolidone (NMP), which was expected to dissolve binders in the cathode material and achieve high efficiency [10], was tested to assess their performance. Among these solvents, distilled water demonstrated the highest efficiency, achieving a 69.4% removal rate of cathode and a 72.6% efficiency of removal for anode material. However, challenges such as reduced efficiency due to sample overlap and limited solvent interaction underscore the need for process optimization, including improved agitation and optimized flow control.

II. Methodology

A. Materials

Electrode samples were extracted from three pouch cells of nickel-manganese-cobalt (designated as NMC622), dismantled from one fully electric battery electric vehicle (BEV) module [11], with the specifications detailed in Tab. I. The aluminium-polymer casings of the deeply discharged cells (with a voltage of approximately 0 V) were carefully opened using a ceramic knife, allowing for the separation of the individual electrode layers. Each cell contained 18 cathode sheets and 19 anode sheets, each of which consisted of a current collector covered on both sides with active electrode material [11], [12]. In this paper, the terms "cathode" and "anode" refer to the positive and negative electrodes, respectively, in alignment with the discharging process [13].

TABLE I. Investigated Battery Cell Specification

Dimensions - height x width x depth (mm)	8.65 x 100 x 548
Weight (kg)	1.1
Capacity (Ah)	78
Nominal voltage (V)	3.67

Based on the cell structure, the cathodes were initially enclosed within layers of separator, which were thoroughly removed to expose the electrode surfaces [11]. After this, 2 × 2 cm samples were cut from both electrode materials (see Fig. 1), which were prepared for further treatment. The material specifications and structural characteristics of the treated materials are provided in Tab. II [12].

TABLE II. Battery Cell Material Characterization [12]

Cathode, double-sided active layer with collector (µm)	~ 189
Cathode active layer, single-sided (µm)	~ 87.5 ($LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ + binders)
Cathode current collector (µm)	~14 (Aluminium)
Anode, double-sided active layer with collector (µm)	~ 243
Anode active layer, single-sided (µm)	~ 115.5 (Graphite + binders)
Anode current collector (µm)	~12 (Copper)

Samples were then thermally treated at 650 °C for 2 hours in a CLASIC chamber furnace and cooled to room temperature [14]. This process eliminated organic components, such as binders and residual electrolytes [15].

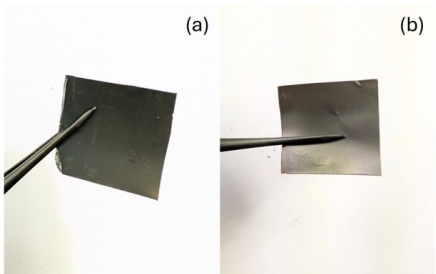

Fig. 1. Illustration of non-thermally treated samples (a) cathode, (b) anode material of lithium-ion pouch battery cell.

B. Ultrasonic Treatment

The calcinated samples were treated with BANDELIN SONOPULS HD 2200.2 ultrasonic homogeniser, with a booster horn SH 213 G, under the same parameters for all experiments (see Tab. III) [16]. To assess the effectiveness of various solvents, experiments were conducted using ethanol, acetone, isopropyl alcohol, NMP, and distilled water. Furthermore, separation efficiency was evaluated not only for individual samples but also for larger samples sets. Experiments were carried out with 1, 10, and 40 specimens placed in the same bath volume to examine the impact of sample quantity on separation. For all experiments, the bath temperature was initially maintained at room temperature.

TABLE III. Ultrasonication Experiments Parameters

Time (min)	Rated Ultrasonic Output (W)	Frequency (kHz)	Amplitude (%)	Bath Volume (ml)
2	200	20	50	60

C. Process Efficiency Evaluation

The efficiency of the process was calculated using the average weight ($m_{electrode}$) per unit area (g/cm²) corresponding to 0.06138 g/cm² for the cathode and 0.03644 g/cm² for the anode materials, as well as the thickness of the current collectors provided in Tab. II. Additionally, the densities of the materials used, i.e., aluminium (2.71 g/cm³) and copper (8.96 g/cm³) [17], were employed to calculate the mass of the collectors. This was achieved using the formula:

$$m_{\text{collector}} = t_{\text{collector}} \cdot A \cdot \rho_{\text{collector}}\ (g), \qquad (1)$$

where $m_{collector}$ represents the mass of the current collector, $t_{collector}$ is the thickness of the collector, A is the electrode area (1 cm²), and $\rho_{collector}$ is the density of the material.

The calculated mass of the current collectors was subtracted from the average sample weight ($m_{electrode}$) to determine the mass of the active electrode materials (m_{active}), i.e., the NMC layer for the cathode and the graphite layer for the anode. This calculation enabled the determination of the percentage distribution of individual electrode components, specifically the active materials and the current collectors, as presented in Tab. IV.

TABLE IV. Electrode Material Distribution

Electrode Material	Active Material Layer, m_{active} (%)	Current Collector Material, $m_{collector}$ (%)
Cathode	93.82	6.18
Anode	70.49	29.51

During the measurements, the weights of the individual samples were recorded both before ($m_{initial}$) and after ($m_{treated}$) the separation process. Based on the data presented in Tab. IV, the proportions of m_{active} were determined. This calculated value represents the maximum amount of material that could be separated from the electrodes.

The separation efficiency ($\eta_{separation}$) was then calculated using the formula:

$$\eta_{\text{separation}} = \frac{m_{\text{initial}} - m_{\text{treated}}}{m_{\text{active}}} \cdot 100\ (\%). \qquad (2)$$

This calculation provided a clear metric, enabling an evaluation of process performance.

III. Results and Discussion

The study's findings were separated into two key parts to provide a comprehensive evaluation. The first part is dedicated to the experimental investigation, which is focused on the efficiency of ultrasonic treatment in separating active materials from electrodes, providing insights into the influence of process parameters on separation performance. The discussion includes material recovery rates, separation efficiencies, and process consistency to demonstrate the feasibility of this method for application in battery recycling.

The second part evaluates the practical considerations and limitations of scaling up the proposed method to an industrial context. This analysis addresses challenges such as material throughput, energy consumption, process integration, and the economic viability of implementing ultrasonic-assisted separation at scale. This section highlights critical barriers and opportunities for advancing the laboratory-scale findings toward real-world applications.

A. Ultrasonic-Assisted Electrode Material Separation

For the samples used within a 1-sample per bath experiment setup, the average sample weight for the cathode material was 0.2455±0.0026 g, with 0.2303±0.0024 g of active and 0.0152±0.002 g of current collector material. For the anode material, the average sample weight was 0.1458±0.0015 g, with 0.0430±0.0004 g of active and 0.1028±0.0011 g of current collector material. The standard deviations for both electrode materials were minor, indicating minimal variations. The details are shown in Tab. V.

TABLE V. Initial Mass Analysis, 1 Sample/Bath

Cathode Material			
Experiment number (1 sample per bath)	***Sample weight (g)***	***Active material (g)***	***Current collector material (g)***
1	0.2438	0.2287	0.0151
2	0.2486	0.2332	0.0154
3	0.2473	0.2320	0.0153
4	0.2456	0.2304	0.0152
5	0.2423	0.2273	0.0150
Average	0.2455	0.2303	0.0152
Standard Deviation	±0.0026	±0.0024	±0.0002
Anode Material			
Experiment number (1 sample per bath)	***Sample weight (g)***	***Active material (g)***	***Current collector material (g)***
1	0.1454	0.0429	0.1025
2	0.1437	0.0424	0.1013
3	0.1476	0.0436	0.1040
4	0.1453	0.0429	0.1024
5	0.1468	0.0433	0.1035
Average	0.1458	0.0430	0.1028
Standard Deviation	±0.0015	±0.0004	±0.0011

The weights of samples and individual materials from the other experiments (10 samples per bath and 40 samples per bath setups) are presented in Tab. VI. As the number of samples increases, the standard deviation also increases, though it remains relatively low, with a maximum of 3.68%.

TABLE VI. Initial Mass Analysis, 10 and 40 Samples used per Bath

Cathode Material			
Experiment number (10 samples per bath)	***Sample weight (g)***	***Active material (g)***	***Current collector material (g)***
1	2.3183	2.1750	0.1433
2	2.3927	2.2448	0.1479
3	2.4942	2.3400	0.1542
Average	2.4017	2.2533	0.1485
Standard Deviation	±0.0883	±0.0828	±0.0055
Anode Material			
Experiment number (10 samples per bath)	***Sample weight (g)***	***Active material (g)***	***Current collector material (g)***
1	1.4369	0.4240	1.0129
2	1.4138	0.4172	0.9966
3	1.4060	0.4149	0.9911
Average	1.4189	0.4187	1.002
Standard Deviation	±0.0161	±0.0047	±0.0113
Cathode Material			
Experiment number (40 samples per bath)	***Sample weight (g)***	***Active material (g)***	***Current collector material (g)***
1	9.7521	9.1493	0.6028
2	9.8018	9.1959	0.6059
3	9.7983	9.1927	0.6056
Average	9.7841	9.1793	0.6048
Standard Deviation	±0.0277	±0.0260	±0.0017
Anode Material			
Experiment number (40 samples per bath)	***Sample weight (g)***	***Active material (g)***	***Current collector material (g)***
1	5.8162	1.7161	4.1001
2	5.7027	1.6826	4.0201
3	5.8964	1.7398	4.1566
Average	5.8051	1.7129	4.0922
Standard Deviation	±0.0973	±0.0287	±0.0686

The minimal variation observed in both cathode and anode materials can be attributed to the precision of the measurements and the samples' homogeneity. However, it is important to note that differences in mass could also result from electrolyte residues accumulating on the electrode surfaces after the thermal treatment, as shown in Fig. 2.

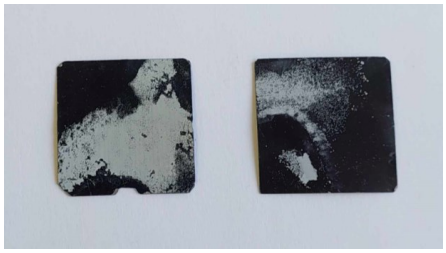

Fig. 2. Electrolyte residues (white) on the thermally treated cathode material samples.

Figure 3 (a) presents the efficiency of electrode material separation using various solvents. Distilled water exhibited the highest efficiency among the tested solvents, achieving approximately 69.4% and 72.6% removal for cathode and anode materials, respectively. Ethanol also demonstrated relatively high efficiency (21.1% for cathode and 34.8% for anode), followed by acetone and isopropyl alcohol, which showed moderate performance. However, despite expectations based on previous studies [6], [8], [9], treated materials had lower removal efficiency for both tested electrode materials. This contrasts with earlier findings where nearly complete cathode and anode material removal was reported. A significant discrepancy in results is evident even when using similar processing parameters, suggesting that additional influencing factors must be considered. A key factor may be differences in electrode compositions, which can significantly impact solvent-material interactions. Nevertheless, a detailed material analysis of electrode compositions would be necessary to clarify these differences.

Beyond its high separation efficiency, distilled water presents a significant advantage over organic solvents due to its environmental compatibility and safety. Unlike ethanol, acetone, or NMP, which are volatile and often require special handling due to toxicity and flammability concerns [18], water is non-toxic, non-flammable, and does not pose disposal challenges [19]. Additionally, using distilled water supports sustainability goals by minimizing chemical waste and reducing the need for energy-intensive processes [19]. This makes it a highly favourable option for large-scale recycling applications, particularly in contexts where safety and environmental impact are primary considerations.

Furthermore, the standard deviations indicate the consistency of the separation process. Distilled water showed high efficiency but exhibited relatively low deviation, suggesting stable performance across trials. In contrast, some solvents, such as isopropyl alcohol, exhibited higher deviation, which may indicate variability in interaction with electrode coatings.

Figure 3 (b) evaluates the effect of sample quantity on separation efficiency using distilled water. The results reveal a clear trend: as the number of processed samples increases from 1 to 40, the efficiency of material separation decreases for both electrode types. Specifically, cathode material removal drops from 69.4% (1 sample) to 32.8% (40 samples), while anode material separation declines from 72.6% to 35.7%. This decrease is likely caused by sample overlap, which reduces solvent accessibility to the electrode surfaces. It can be expected that for every separation, the efficiency will be stabilized at a certain threshold, as some degree of material separation will always occur due to mechanical and chemical interactions within the solvent medium. However, in practical or industrial applications, additional steps, such as agitation or flow control, would be implemented to enhance sample circulation, minimize overlap, and increase efficiency.

Both electrode materials show similar separation trends, indicating that the applied method does not significantly differentiate between the cathode and anode for this cell type. This implies that the separation process is mainly influenced by physical constraints, such as particle adhesion and limitations in solvent diffusion, rather than by intrinsic differences in electrode composition. Since the materials have undergone thermal treatment, it is expected that the polymeric binders have been completely removed. Therefore, the differences in separation efficiency are likely not due to binder dissolution but rather to variations in material adhesion and the structural integrity of the electrode layers. However, it is possible that residual traces of the binder or its decomposition products remain in the material, which could contribute to the observed differences in detachment efficiency. Additionally, strong interfacial interactions between the active materials and the current collectors, possibly affected by the thermal processing conditions, could further impede efficient separation. As a result, while this method shows comparable efficiency for separating the cathode and anode in the tested system, variations in material bonding, electrode microstructure, or any remaining binder residues in other battery chemistries might lead to different outcomes.

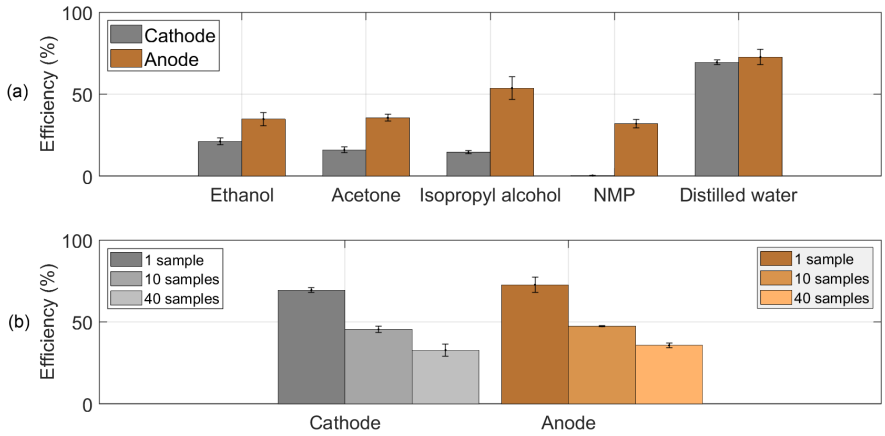

Fig. 3. The efficiency of lithium-ion battery pouch cell electrode ultrasonic-assisted material separation under different processing conditions. (a) Comparison of efficiencies using different solvents. NMP = N-Methyl-2-pyrrolidone. (b) The influence of the number of samples per bath on the separation performance for the experiments using exclusively distilled water.

B. Industrial-Scaled Process Application Challenges

Although the presented laboratory-scale results provide valuable insights into the separation efficiency of electrode materials, several factors must be considered and carefully addressed before transitioning to industrial applications. Key considerations include maintaining process efficiency at higher throughput, ensuring material purity, and minimizing environmental impact. Tab. VII summarizes the main obstacles and potential solutions that should be considered for successful large-scale implementation.

TABLE VII. INDUSTRIAL-SCALE APPLICATION CHALLENGES OF ULTRASONIC-ASSISTED ELECTRODE MATERIAL SEPARATION

Consideration	Challenge	Description
Ensuring of Process Efficiency	Reproducibility & Scale-Up	Maintaining consistent separation across large volumes while maximizing solvent penetration and agitation.
	Processing Time Optimization	Balancing separation efficiency with material integrity while adapting processes for industrial throughput.
Maintenance of Material Purity	Material Purification & Filtration	Implementing filtration or sedimentation steps to eliminate fines and prevent inefficiencies in the process.
	Separation of Recovered Fractions	Guaranteeing high purity and uniformity of recovered electrode materials for reuse.
Minimization of Environmental Impact	Waste Management & Solvent Recovery	Developing strategies for solvent recycling and minimizing residual contamination.
	Regulatory & Environmental Compliance	Adjusting waste treatment and recovery processes to meet industrial and environmental standards.

One major challenge is the reproducibility on a larger scale. While working with small batches allows for precise control over various parameters, scaling up introduces issues such as uniformity, solvent penetration variations, and agitation challenges. Moreover, the selectivity of the separation process can differ based on the specific type of battery and its binder composition, which ultimately affects the overall efficiency. Process time is another important factor that must be considered to maintain process efficiency. Although this study doesn't explicitly detail time-dependent experiments, additional trials have revealed that a two-minute treatment period is optimal for achieving the highest separation efficiency. Shorter exposure times tend to result in lower efficiency, while longer treatments can damage current collectors and contaminate the recovered active materials. In industrial applications, balancing these factors is crucial. While the optimal time might not vary significantly between laboratory and large-scale processing, achieving high efficiency would necessitate engineering adjustments, such as increasing the number of probes or fine-tuning their distribution within the processing bath.

Achieving efficient separation is just one aspect that must be considered within the broader framework of industrial operations, including recovery and purification management, which presents another critical challenge for large-scale applications. Even with high separation efficiency, it is essential to ensure the effective filtration and classification of the recovered fractions. In a scaled-up system, the accumulation of fine particulate matter and binder residues can lead to clogging or reduced process efficiency. This situation necessitates the integration of filtration steps or sedimentation processes to maintain operational stability. Furthermore, controlling the quality and consistency of the recovered materials is vital, as variations in purity can affect their reuse in battery manufacturing, particularly regarding the application of active materials, and in secondary applications, such as the use of current collector materials.

Furthermore, waste handling and environmental considerations must be addressed when implementing ultrasonic-assisted separation on an industrial scale. One advantage of the presented approach is using water as the primary processing medium, reducing the need for hazardous organic solvents and contributing to a more environmentally friendly process. Additionally, the prior thermal treatment of electrode materials significantly reduces the binder content, facilitating cleaner separation. However, despite these advantages, the complete elimination of residual contamination remains challenging. Even with optimized processing conditions, trace amounts of degraded binders and fine particulate residues may still affect the purity of the recovered active materials. Therefore, large-scale implementation would necessitate solvent recovery strategies, fine filtration systems, and optimized separation steps to ensure product quality while minimizing waste. Depending on the regulatory framework, additional treatment for residual binders and metal impurities may be necessary to comply with industrial and environmental standards. As such, the scalability of this method must include not only efficiency improvements but also robust strategies for solvent recycling, waste minimization, and resource recovery to create a sustainable and economically viable process.

IV. CONCLUSION

As the demand for LIB recycling increases, it is essential to develop efficient and sustainable methods for material recovery. One promising approach represents ultrasonic-assisted electrode material separation, which reduces the need for hazardous solvents while maintaining high separation efficiency. This study shows that the choice of solvent and the processing conditions significantly affect the efficiency of material separation. Distilled water emerged as the most effective and environmentally friendly option, outperforming traditional solvents and eliminating concerns related to toxicity and flammability.

However, the study also identified challenges related to sample processing in the ultrasonic-assisted separation process. High efficiency was achieved when processing single samples but increasing the number of samples per batch resulted in a noticeable decline in separation performance due to sample overlap and limited solvent interaction. This finding highlights the need for process optimization in large-scale operations. Potential solutions include adding agitation mechanisms, adjusting bath configurations, or implementing solvent flow systems to enhance sample exposure and maintain separation efficiency.

Future research should investigate the role of material adhesion mechanisms and potential residual binder residues on solvent-material interactions on separation efficiency. Additionally, a thorough analysis of the scalability of the ultrasonic-assisted separation process is necessary to connect laboratory-scale testing with industrial recycling applications.

ACKNOWLEDGMENT

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Estimation of Material Flows during Recycling of Waste Lithium-Ion Batteries from Electric Vehicles in the Czech Republic

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Abstract. *Lithium representation in battery technology is undeniable, and its application shows in decades an extending trend. Lithium-ion batteries (LIBs) play the most crucial role in energy storage systems, powering consumer electronic devices, and even electric vehicles. At the same time, there is a breaking point in the entire automotive industry; increasing requirements for emission reductions are hampering the new production of combustion vehicles and rising demand for electric ones. The rapid global growth in the electric vehicles (EVs) market brings numerous waste-management challenges in end-of-life treatment, mainly devoted to remarkable differences between production and recycling rates.*

In this paper recycling flows for two different lithium-ion technologies of waste batteries from EVs (LMO, NMC) discarded between 2020-2025 in the Czech Republic (CZ) were estimated. The results present the weights of recovered materials from three different EV types (Battery Electric Vehicle - BEV, Plug-in Hybrid Electric Vehicle - PHEV, Hybrid Electric Vehicle - HEV), analyze the gradual recycling of the battery pack-module-cell, and compare two scenarios of used recycling rates.

Keywords

Lithium-ion batteries, electric vehicles, recycling processes, material flows, the Czech Republic.

1. Introduction

Electric vehicles (EVs) fully meet the ever-increasing requirements of a reduced environmental impact compared to conventional combustion vehicles. These vehicles in purely electric mode produce zero tailpipe emissions, lower smog, and lower greenhouse gas emissions [1]. Therefore, it is assumed that 20% of all cars sold worldwide will be electric by 2025 [2]. In the EU, there will be about 4 million registered EVs in the mentioned year, and this number will continue to rise [3].

Lithium technology is dominant in the electric automotive industry. Lithium-ion batteries (LIBs) are typically based on lithium (Li), cobalt (Co), manganese (Mn), and nickel (Ni) in the cathode, graphite (C) in the anode, aluminum (Al), copper (Co), and iron (Fe) in other components. Their composition affects properties, such as energy density, rechargeability, or cycle life. Commonly used LIBs according to their cathode material are for example lithium-nickel-cobalt-manganese oxide (NMC), lithium-manganese oxide (LMO), and lithium cobalt oxide (LCO) [2], [4], [5].

Due to the fast growth of the EV market, the issue of waste batteries disposal must be solved. The EU legislation supports the idea of recycling processing embodied in the EU Battery Directive (currently, at least 50 % of waste batteries weight must be recycled) [6]. Recycling is a suitable solution [4], [6], but it is necessary to understand the flows composition of input (waste) and output (recovered) materials. Prediction of the weights of obtained materials can be used to improve existing recycling plants, modify the design of new ones, or reduce the extraction of raw materials when using recovered materials in new production.

In this paper, the model for estimation of recycling material flows for two types of lithium-ion batteries (LMO, NMC) and three models of EVs (Battery Electric Vehicle - BEV, Plug-in Hybrid Electric Vehicle - PHEV, Hybrid Electric Vehicle - HEV) was developed. Energy sources of these EVs are schematically shown in Fig. 1. The developed model performs weight prediction for battery materials (Co, Li, Mn, Ni, C, Al, Cu, Fe) during 2020-2025 for two recycling scenarios in the Czech Republic (CZ).

Fig. 1. Energy sources of electric vehicles – BEV, PHEV, HEV.

A. Pražanová, and Z. Plachý, “Estimation of Material Flows during Recycling of Waste Lithium-Ion Batteries from Electric Vehicles in the Czech Republic” [C7], © 2022 by the authors.

2. Determination of the model

The proposed model is based on studies of the EVs market and proposed waste-management solutions [3]–[5], [7], [8] which were analyzed in the conditions of the Czech Republic (CZ). The schematic structure of developed model is illustrated in Fig. 2.

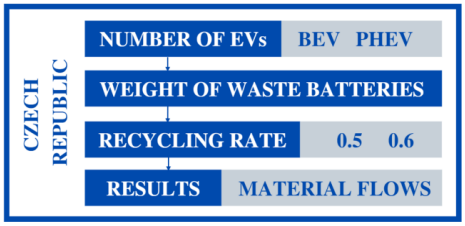

Fig. 2. The structure of developed model.

2.1 Number of EVs in the Czech Republic

The estimation of the number of EVs in CZ was based on data Ministry of Industry and Trade, Ministry of Transportation, Czech Republic [7] of registered electric vehicles during the years 2015-2020. These data are shown in Fig. 3, where the total number of EVs and their distribution into BEVs and PHEVs is visible.

A second-degree polynomial was used to extrapolate the number of EVs in 2020-2025. The EVs estimate, including their type of distribution in CZ in 2020-2025, is demonstrated in Fig. 3.

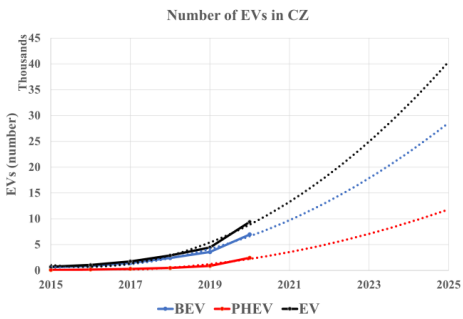

Fig. 3. Number of EVs in CZ in 2015-2025, dotted = estimated.

2.2 Recycling rate

The minimum recycling rate for waste batteries is determined by EU Battery Regulations. The current requirement is that at least 50 % of a waste battery’s weight must be recycled. From 2025, this requirement will increase to 65 % and up to 70 % from 2030 [6].

2.3 Flows of recycling materials

The estimation of discarded batteries in CZ in 2020-2025 was based on case studies of Urban Mine Platform [9]. This case study considers the BEV, PHEV, and HEV models in CZ (quantity proportion 4:2:1) of the chemical composition LMO and NMC (proportion 4:3) [9].

The weights and compositions of individual components (battery pack, module, cell) of LMO and NMC batteries were determined for the mentioned models of electric vehicles from the study by Argonne [5]. The considered composition and weight of individual components for the LMO BEV battery are listed in Tab. 1.

Battery composition, weight of components (kg)		
Pack (49.14)	Module (15.68)	Cell (317.95)
Copper (0.08)	Copper (0.39)	Active Cathode Material (166.35)
Aluminum (33.48)	Aluminum (13.94)	Graphite (56.19)
Steel (2.23)	Plastic (0.13)	Carbon (3.47)
Insulation (1.06)	Insulation (0.1)	Binder (PVDF) (4.61)
Coolant (7.92)	Electronics (1.12)	Copper (31.02)
Electronics (4.36)		Aluminum (17)
		Electrolyte (34.9)
		Plastic (4.42)

Tab. 1. Composition of individual battery components.

Subsequently, the waste batteries' materials were determined; LMO was considered 1:2 (Li:Mn), and NMC 1:3:3:3 (Li:Ni:Mn:Co) [5]. According to current legislation, a minimum part of the total weight of these batteries was selected for recycling [6].

The model works with gradual and separate recycling of individual components. Firstly, the battery pack is dismantled, the covering plastic and metal materials are taken off. Secondly, the modules covering, and electronics are removed. Finally, the battery cell, including the active material, is recycled. These recycling steps are performed separately, and their efficiency is summarized in Tab. 2 [4].

Elements	Recycling efficiency [-]		
	Pack	Module	Cell
Al	0.9	0.9	0.8
Cu	0.9	0.9	0.8
Co	-	-	0.85
C	-	-	0
F	-	-	0
Fe	0.9	0.9	-
Li	-	-	0
Mn	-	-	0.95
Ni	-	-	0.95
P	-	-	0

Tab. 2. Recycling efficiency of individual recycling steps.

3. Estimation of material flows

The results of the developed model were divided into three parts. The evaluation of the weights of input and output materials of BEV, PHEV, HEV models in CZ in 2020-2025; the comparison of output materials from recycling the individual components pack-module-cell; the observation of change when changing recycling rate from 50 to 65 % in 2025 due to the valid legislation.

3.1 Material flows from EV batteries

The input and output materials quantities during recycling are shown in Fig. 4-6. These figures describe materials flows for BEVs, PHEVs, HEVs, during 2020-2025, respectively.

The material flows respect the quantity distribution of models on the EV market, in descending order BEV, HEV, PHEV. The most discarded batteries were recorded from the BEVs, where the weight of waste batteries (input) was ~70 tonnes; approximately 40 tonnes were recovered.

Estimated material flows respect the chemical composition of individual batteries and their purpose of application. The proportion of active material, especially manganese (Mn), in BEVs is higher than in other models. This EV type is fully electrical and cannot be powered in any other way. Thus, the high energy and power requirements must be satisfied. For the rest of the models, PHEV and HEV dominate the weight of covering materials, notably aluminum (Al).

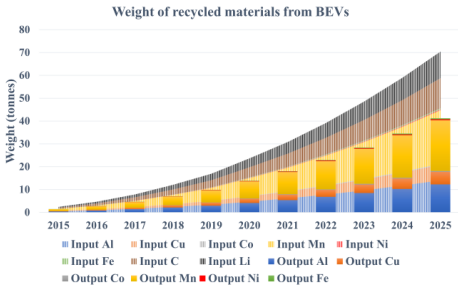

Fig. 4. Weight of recycled materials (input/output) - BEVs.

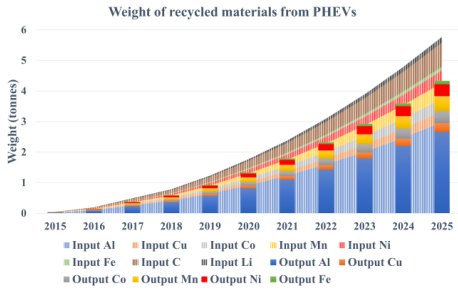

Fig. 5. Weight of recycled materials (input/output) - PHEVs.

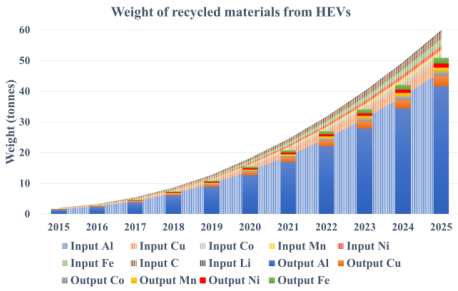

Fig. 6. Weight of recycled materials (input/output) - HEVs.

3.2 Battery pack-module-cell materials

Fig. 7 compares recycled materials (outputs) of individual components of LMO, and NMC waste batteries from BEVs in 2021. This procedure corresponds to the established recycling processes in the EU. Therefore, there is no lithium (Li) recovery. Graphite (C) is lost during high-temperature processing; it is used as a reducing agent).

From the composition of the output materials of individual components, their primary function is obvious: the battery cell provides the electrochemical part and consists of valuable metals; the module and pack provide coverage and protection with common metals.

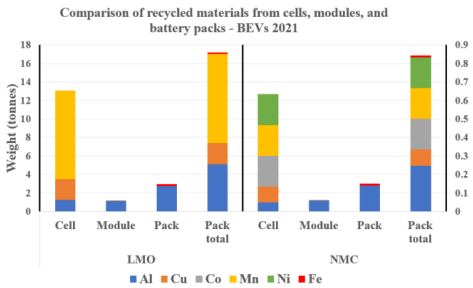

Fig. 7. Comparison of recycled materials (output) from battery cells, modules, and packs in 2021.

3.3 Change in regulation, change in rate

In 2025, the mandatory recycling rate stated by the waste batteries' weight would be increased from 50 to 65 %. This change is simulated in Fig. 8. The increase in recovery is reflected in all recovered materials, mainly aluminum and manganese. The 15% increase in the recycling rate will affect the weight of recovered materials; there will be an increase of approximately 25 tonnes.

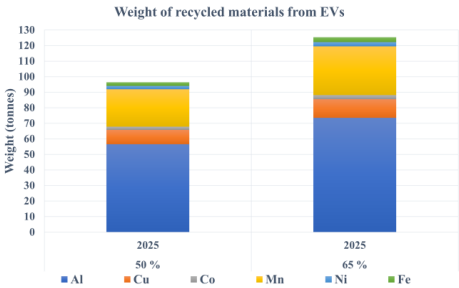

Fig. 8. Comparison of quantity of recycled materials (output) from EVs using recycling rate 50 %, and 65 % in 2025.

4. Conclusion

This work estimates material flows from waste electric vehicles lithium batteries in the Czech Republic in 2020-2025. The obtained results describe the current issues of electromobility in the conditions of the Czech Republic and can be used while designing a new recycling factory or solving waste-materials logistics.

In the Czech Republic, an approximately 2.5 times higher ratio of BEV to PHEV in 2020-2025 is expected. In 2025, the total number of electric vehicles should reach the limit of 40,000.

With the growing number of electric cars in the Czech Republic, waste materials from discarded batteries will increase. The tightening of legislation and the increase in the recycling rate from 50 to 65% in 2025 should increase the recycled material obtained by 25 tonnes.

This model will be extended with an economic and environmental view, such as costs or emissions related to transport to the recycling plant and recycling process implementation as part of future work.

Acknowledgements

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43. NEKONVENČNÍ ZDROJE ELEKTRICKÉ ENERGIE

EKONOMICKÁ NÁROČNOST RECYKLACE LITHIUM-IONTOVÝCH BATERIÍ Z ELEKTROMOBILŮ

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Abstract

Emissions reduction has been tightened worldwide, especially in automotive. Electric vehicles (EVs) are a suitable solution that can meet the strict requirements in CO_2 production and high user torque and speed requests. Therefore, a significant increase in demand for EVs has occurred. This growing trend results in increased production of new vehicles, raw materials consumption, and the number of waste vehicles and batteries on the market. Solution methods are being sought: the recycling process is one of the most promising.

This work provides a simplified overview of the economic evaluation of the recycling process of spent lithium-ion batteries from EVs in conditions of the Czech Republic. The described technique evaluates a combination of the pyrometallurgical and hydrometallurgical methods (with the process efficiency above 95 %) and the high quality of output products. Moreover, this work presents future scenarios considering the changes due to EU legislation.

Úvod

Nároky na snižování produkce emisí v automobilovém průmyslu se stále zvyšují, především kvůli cílům Zelené dohody pro Evropu (*European Green Deal*) a balíčku „Fit for 55“. Mezi vhodná a uživatelsky přívětivá řešení se řadí elektromobily, motorová vozidla na elektrický pohon, které pro ukládání energie využívají baterie založené především na lithium-iontové technologii. Tyto vozy vyhovují vysokým požadavkům na produkci CO_2 během provozu a uživateli zaručují dostatečný komfort v podobě dojezdových vlastností, eliminace nadměrného hluku nebo vlivu na lokální znečištění [1].

Rostoucí počet elektromobilů na trhu, který byl zaznamenán v minulých letech, by jen v rámci EU mohl k roku 2030 dosáhnout 30 milionů čistě elektrických vozů (současně je v EU registrováno méně než 3 miliony těchto vozů) navyšuje spotřebu surových materiálů používaných při výrobě baterií. Lithium-iontové baterie jsou převážně tvořeny cennými kovy jako je kobalt (Co), nikl (Ni), mangan (Mn), lithium (Li) a dalšími prvky jako je grafit (C), hliník (Al), měď (Cu) a železo (Fe). Vhodným řešením, které snižuje spotřebu těchto materiálů, energetickou zátěž a vznik skleníkových plynů během těžby je recyklace [2].

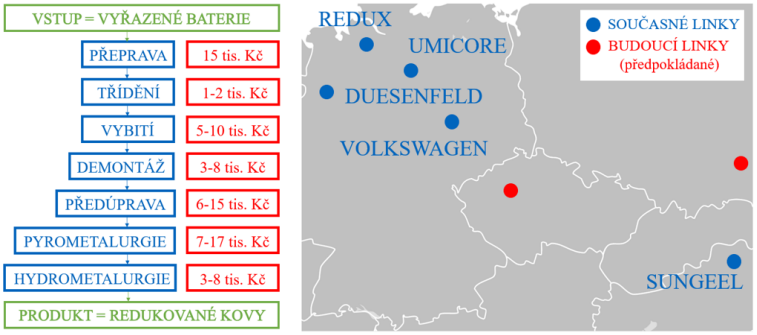
Samotný proces recyklace je podmíněn platnou legislativou (Směrnicí Evropského parlamentu a Rady 2006/66/ES), který stanovuje povinnost výrobci zpětně nashromáždit 95 % dodaných baterií na trh a 50 % z jejich celkové hmotnost recyklovat. Evropská komise vydala předpis, který povinnou míru recyklace baterií navyšuje a to na 65 % k roku 2025 a na 70 % k roku 2030 [1].

Recyklačním procesem, sestávající z několika vhodně vybraných kroků, jsou znovuzískány především aktivní materiály elektrod a kovy obalových, krycích a elektrotechnických materiálů. V současnosti k nejvyužívanějším procesům zpracování vyřazených baterií patří pyrometalurgická a hydrometalurgická metoda, dosahující účinnosti procesu více než 90 % (pyrometalurgie) a 95 % (hydrometalurgie). Ekonomie procesů je vždy závislá na používané technologii a principu a požadavkům na výstupní produkt [2], [3].

Tato práce přináší zjednodušený přehled ekonomické náročnosti jednotlivých kroků recyklace vyřazených lithium-iontových baterií z elektromobilů na území ČR. Zhodnocen byl proces kombinované pyrometalurgické a hydrometalurgické metody, která při současném zavedení dosahuje vysoké účinnosti procesu (nad 95 %) s vysokou kvalitou výstupních produktů pro dva typy elektromobilů (malý vůz s kapacitou přibližně 32 kWh a velký vůz s kapacitou přibližně 80 kWh). Práce zahrnuje i scénáře vývoje a změny cen v důsledku zavedení nově platné legislativy v časovém horizontu 5 a 10 let. Výsledky mohou být užity např. pro výpočet návratnosti investice (*Return on Investment*) při projektu nové recyklační linky.

Experiment

Výše finančních prostředků, která musí být na recyklování lithium-iontových baterií vynaložena je přímo ovlivněna technologickou účinností celého procesu a kvalitou zpětně získaných produktů. Rostoucí účinnost využitých technologií vede ke snižování ztrát celého procesu. Dále pak získání vysoce kvalitních výstupních produktů vede ke zvýšení ekonomických aspektů recyklační linky např. snížení doby návratnosti počáteční investice. Ekonomická náročnost recyklačního zpracování jedné baterie (packu) pro malý a velký vůz (minimální a maximální cena), kombinujícího prvky hydrometalurgické a pyrometalurgické metody, byla stanovena dle schématu uvedeného na Obr. 1 (a) [2].



(a) (b)

Obr. 1: (a) Schéma recyklačního procesu pro stanovení ekonomické náročnosti procesu, (b) mapa současných a budoucích recyklačních linek na území okolních států ČR.

Přeprava odpadních baterií

Recyklace baterií je možná pouze v rámci specializovaných linek, které jsou pro zpětné získaní produktů cenných kovů a ostatních využívaných materiálů vhodně uzpůsobeny. Na území EU se v současné době nachází přibližně 10 vhodných linek, nejvýznamnější z nich situované v blízkosti ČR jsou uvedeny na Obr. 1 (b). Cena přepravy závisí na lokaci sběrného místa odpadních baterií. V současné době jsou pro recyklaci baterií na území ČR vybírány linky na území Německa. V následujících letech lze předpokládat, že budou vybudovány recyklační linky dle Obr. 1 (b). Nové lokace linek celkovou cenu přepravy ovlivní.

Počáteční třídění

Před dalším zpracováním jsou jednotlivé bateriové celky (packy, moduly či články) roztříděny. Prvotní separace probíhá manuálně, zjednodušuje další kroky celého procesu a snižuje bezpečnostní riziko vyřazením porušených komponent [2], [3].

Hluboké vybití

Než bude možné baterie dále zpracovat, je nutné zajistit jejich plné vybití. V opačném případě by při demontáži mohlo dojít ke vzájemnému kontaktu elektrod a zkratu, který generuje vznik Joulova tepla, zapálení těkavých a hořlavých rozpouštědel elektrolytu a nekontrolovatelnému samovznícení celého článku. K vybíjení se především využívá aktivní zátěže s doplňkovou kontrolou celého procesu a rekuperací přebytečné energie do sítě [3].

Demontáž a rozebrání

Z plně vybitých baterií jsou manuálně odebrány obalové a krycí materiály (plasty, kovy) a dochází k odstranění dalších snadno oddělitelných prvků. Tyto materiály jsou přímo dále využívány nebo jsou redukovány ve vysoké kvalitě bez znečištění příměsí jiných prvků [3].

Recyklační předúprava

Před samotnou recyklací jsou vytříděné části baterií, primárně obsahující cenné kovy, vhodně ošetřeny. Části komponent či jejich celé celky jsou drceny. Po separaci materiálů dle velikosti získaných částic drti dochází k termálnímu a chemickému namáhání, které odděluje částice aktivních prvků od proudových sběrnic. Průběžnou separací jsou získávány především kovové materiály, kterou jsou zpracovány dalšími procesy. Vhodná kombinace jednotlivých technik značně ovlivňuje účinnost dále aplikované recyklační metody [3].

Pyrometalurgická metoda

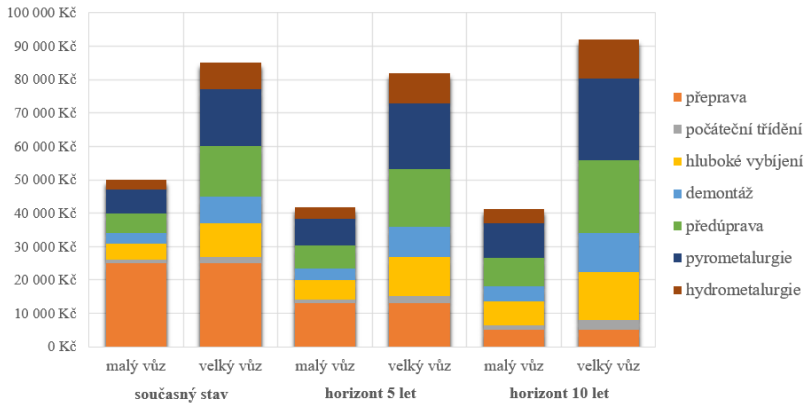
Během pyrometalurgického zpracování dochází k redukci cenných kovů pomocí pražení/kalcinace a tavení ve vysokoteplotní peci. Při nižších teplotách procesu dochází k fyzikálním transformacím (např. fázovým přechodům), při vyšších teplotách jsou inicializovány chemické reakce – jednotlivé materiály se taví. Po roztavení dochází ke koncentrování kovů do slitin na dně pece. Zde mohou být kovy jako Co, Cu, Ni, Al, Mn, či Li struska, s nižší kvalitou produktu, přímo redukovány. Pro docílení vyšší kvality výstupu jsou cenné kovy dále získávány pomocí hydrometalurgických metod [2], [4].

Hydrometalurgická metoda

Celý hydrometalurgický proces probíhá v kapalném prostředí, vhodně zvolenými fyzikálními a chemickými metodami, které zvyšují celkovou výtěžnost aktivních materiálů. Metoda může být využita samostatně, popř. na produkty pyrometalurgie. Vstupní produkty jsou ošetřeny činidly a luhovány v nádržích, které zaručují i separaci nečistot. Cenné kovy jako Co, Li, Mn, Ni a grafit jsou redukovány selektivním srážením či jinými elektrochemickými technikami využitím hydroxidů či sloučenin kovových solí [2], [4].

Výsledky a diskuse

Dle odhadu cen jednotlivých kroků procesu uvedených na Obr. 1 (a) byla stanovena celková (minimální a maximální, dle velikosti kapacity vozu) cena recyklace lithium-iontových baterií z elektromobilů převážených z odběrných míst na území ČR k nejbližším linkám na území Německa. Tato cena uvedená na Obr. 2 jako „současný stav“ odpovídá kombinované pyrometalurgické a hydrometalurgické metodě s účinností nad 95 % s výstupními produkty nejvyšší kvality.



Obr. 2: Ekonomická náročnost recyklačního procesu.

Následně byl odhadován vývoj ceny recyklačního procesu, který nastane v příštích letech. Při uvažování využití polské (za 5 let) či české recyklační linky (za 10 let) dochází ke snížení nákladů na přepravu. Současně je uvažováno roční navýšení účinnosti používané technologie, která snižuje vynaložené náklady v poměru 1:2 vůči odhadovanému ročnímu růstu cen energií. Dosažené výsledky prezentuje Obr. 2 v rámci příslušných horizontů.

Závěr

Elektromobilita představuje již neodmyslitelnou oblast automobilového průmyslu. Počet prodaných elektromobilů na trhu meziročně roste, stejně tak se zvyšuje i počet vyřazených vozů a baterií určených k likvidaci. Materiálové úspory a snížení spotřeby energií vázaných k produkci nových baterií podporuje myšlenku recyklace vyřazených komponent.

Tato práce reflektuje současnou ekonomickou náročnost recyklačního procesu vyřazených lithium-iontových baterií z elektrických vozidel na území ČR. Přiblíženy byly i scénáře dalšího vývoje ceny v důsledku změn legislativy EU vedoucí k výstavbě dalších recyklačních linek a snížení ceny užívaných recyklačních procesů.

Poděkování

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