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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

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.

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.

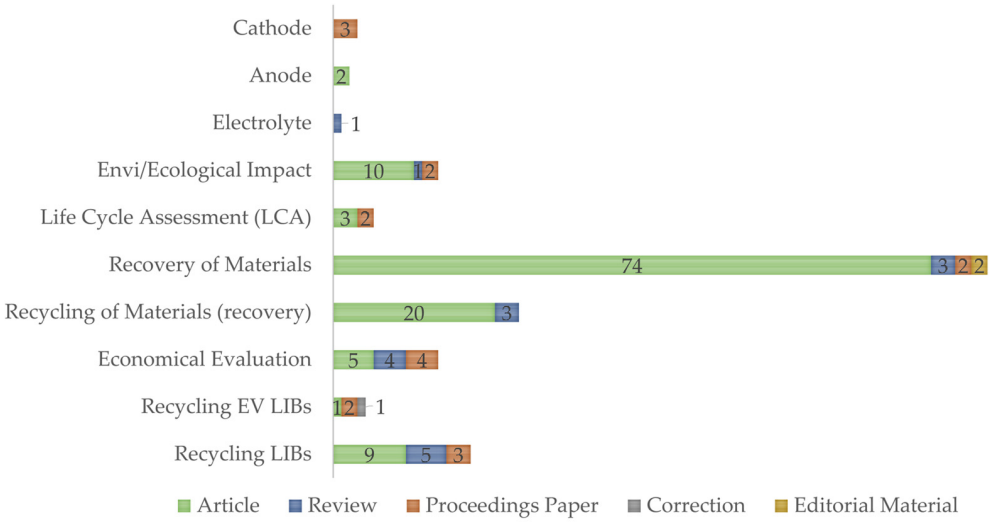


Figure 1. Distribution of reviewed publications according to their type.

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.

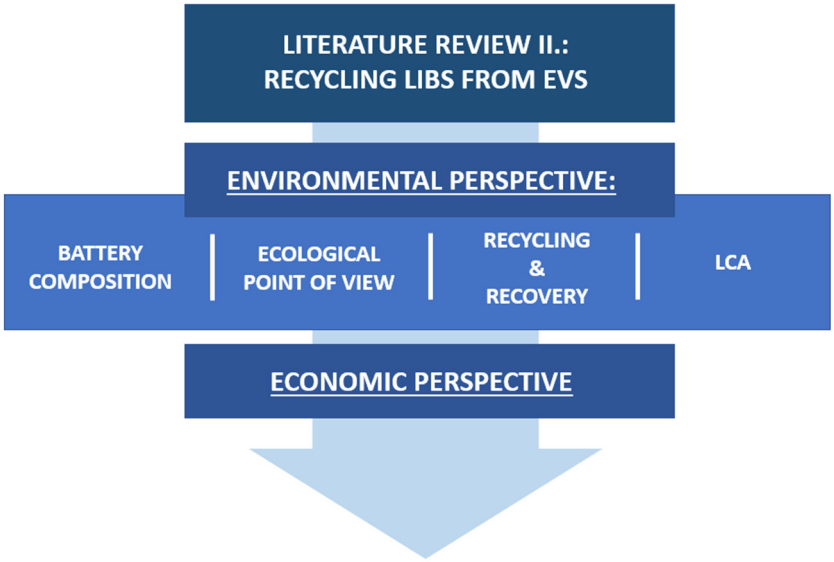


Figure 2. Methodology of the work devoted to E&E of the recycling LIBs from EVs.

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.

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].

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.

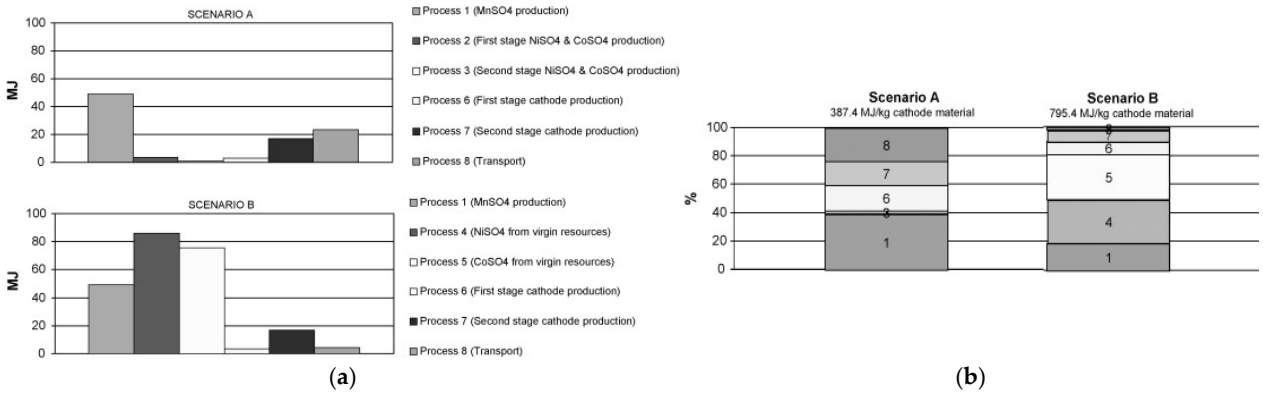
3.1.2. Ecological Point of View

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.



(**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

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

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.

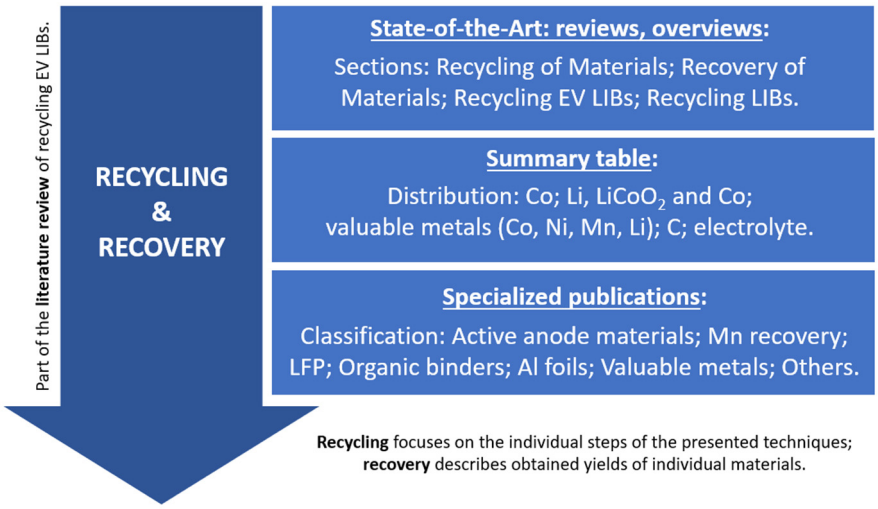


Figure 4. Methodology of Recycling Processes and Recovery Efficiencies caption.

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.

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.

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]

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]

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]

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]

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]

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]

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

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.

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

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

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].

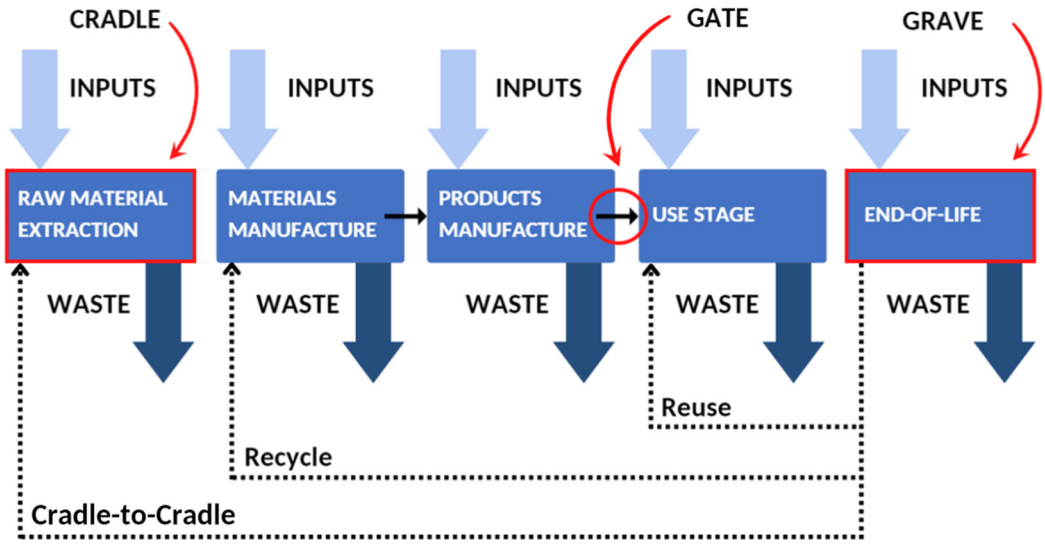


Figure 5. LCA stages diagram including Cradle-to-Grave loop.

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.

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].

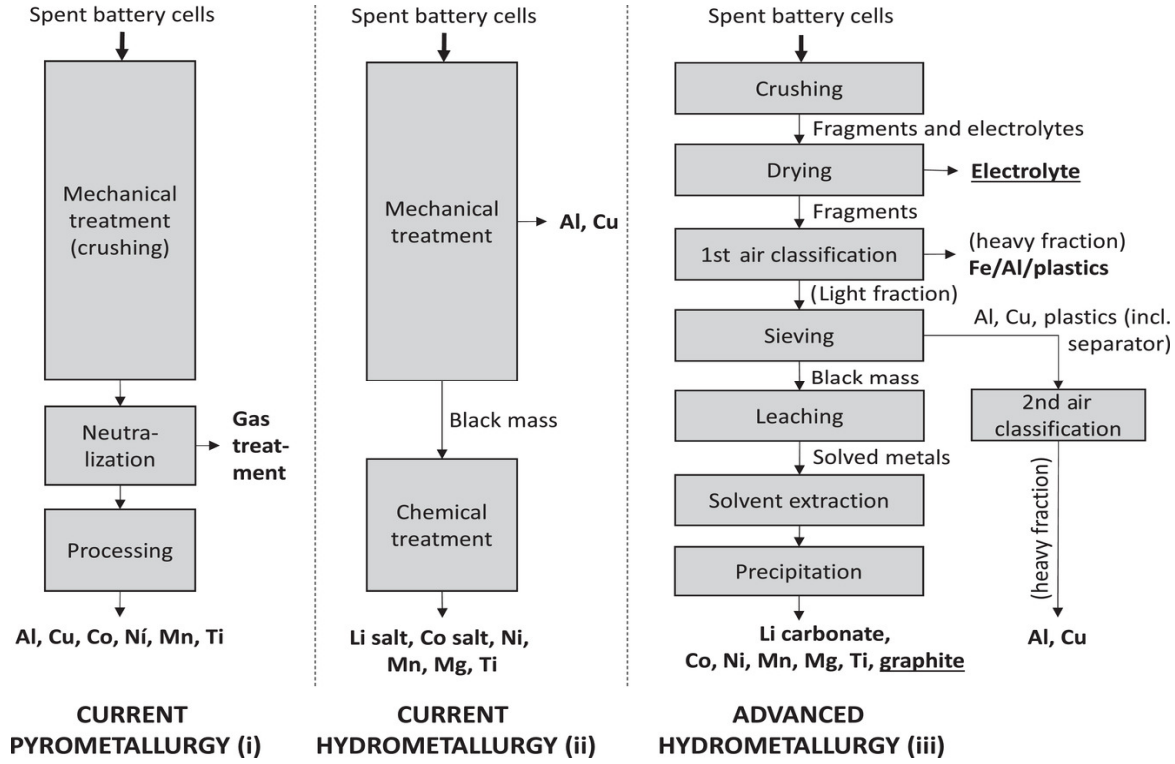


Figure 6. Process flows, including all considered product outputs. Reprinted from Journal of Industrial Ecology, Vol. 24, Mohr et al. [161], Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes, no. 6, pp. 1310–1322, Copyright (2022), distributed under Creative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

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

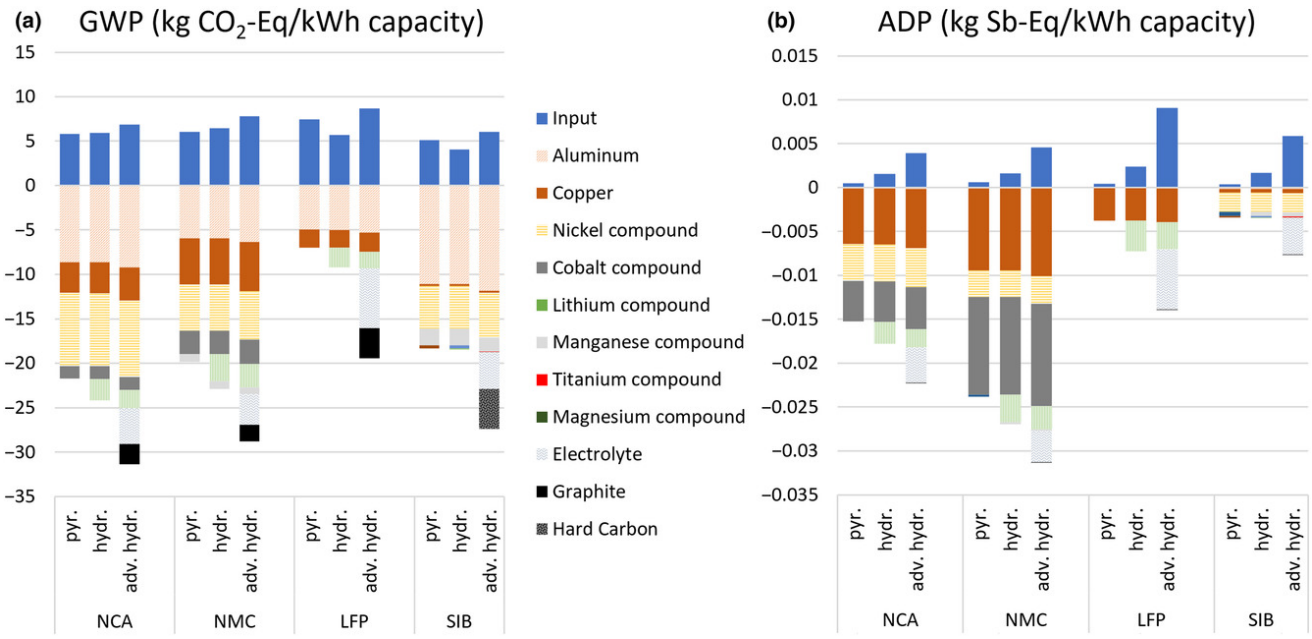


Figure 7. Environmental benefits of battery cell recycling, broken down to the contribution of the different fractions recovered by the recycling processes: (**a**) global warming potential (GWP), (**b**) abiotic resource depletion potential (ADP). Negative values indicate net benefits (reduction of 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

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.

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^{TM}	$CoCl_2$	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.

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.

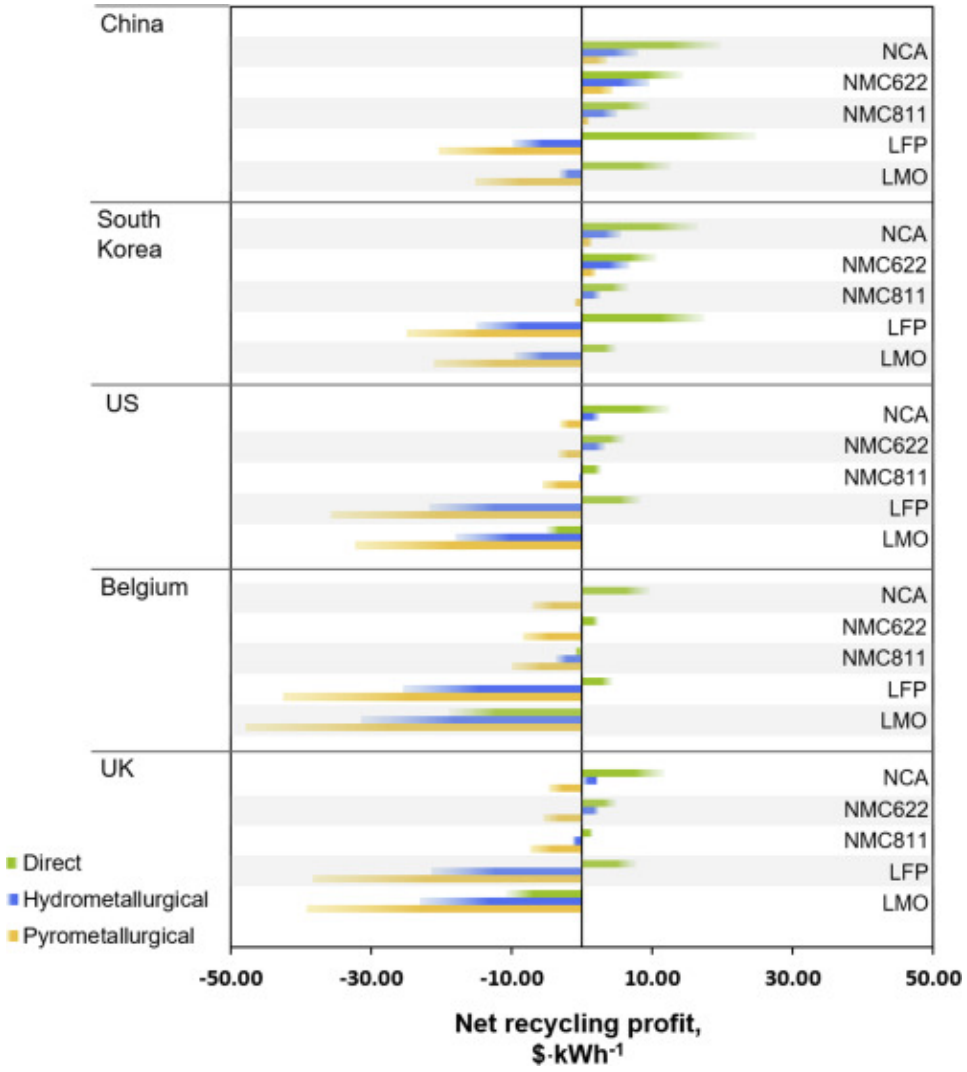


Figure 8. Net recycling profits for commercial EV battery packs. Reprinted from iScience, Vol. 24, Lander et al. [4], Financial viability of electric vehicle lithium-ion battery recycling, no. 7, Copyright (2022), distributed under Creative Commons Attribution License.

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.

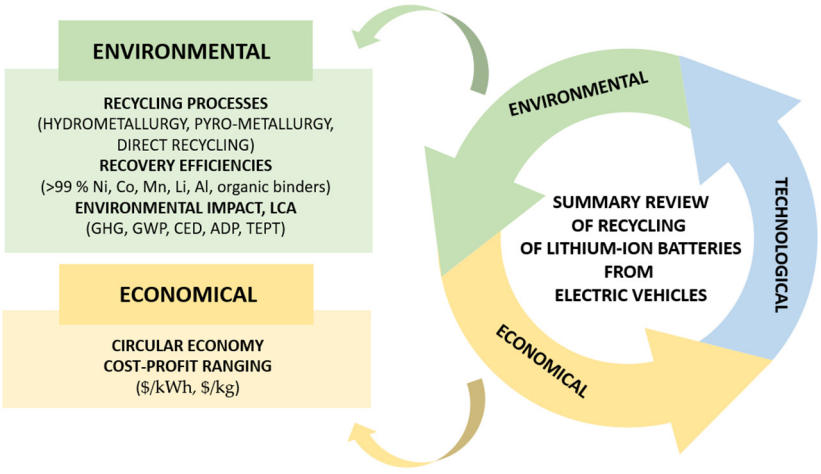


Figure 9. The summarizing figure of the second part of the literature review on recycling lithium-ion batteries from electric vehicles focused on environmental and economic impacts.

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

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.

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.

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.

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.

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.

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.

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.

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.

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.

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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