energies

Review

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

Anna Pražanová [1,*], Vaclav Knap [1,2,3] and Daniel-Ioan Stroe [3]

[1] Faculty of Electrical Engineering, Czech Technical University in Prague, Technická 1902/2, 16627 Prague, Czech Republic; vaclav.knap@cvut.cz
[2] GomSpace A/S, Langagervej 6, 9220 Aalborg, Denmark
[3] Department of Energy, Aalborg University, Pontoppidanstraede 111, 9220 Aalborg, Denmark; dis@energy.aau.dk
* Correspondence: anna.prazanova@cvut.cz

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

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

Citation: Pražanová, A.; Knap, V.; Stroe, D.-I. Literature Review, Recycling of Lithium-Ion Batteries from Electric Vehicles, Part I: Recycling Technology. *Energies* **2022**, *15*, 1086. https://doi.org/10.3390/en15031086

Academic Editor: Haifeng Dai

Received: 19 December 2021
Accepted: 26 January 2022
Published: 1 February 2022

1. Introduction

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

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

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

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

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

2. Review Methodology

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

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

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

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

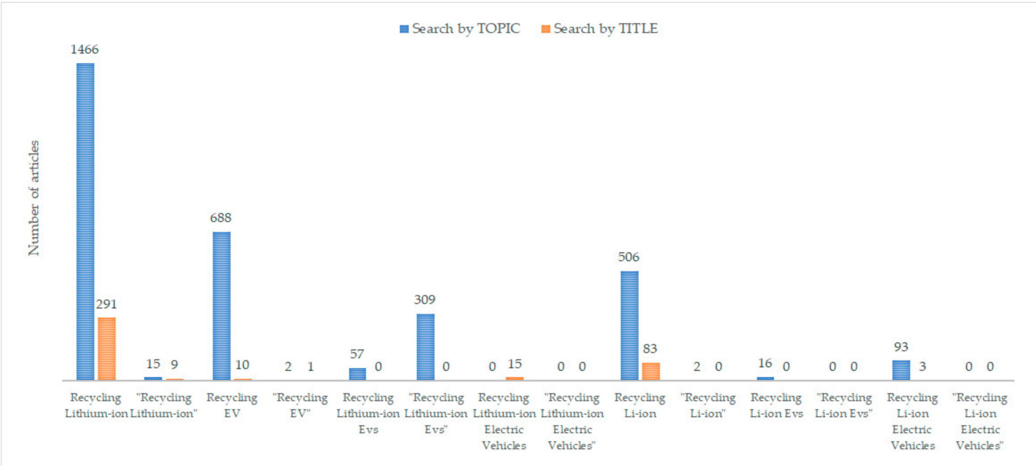

Figure 1. The main search on WoS, the number of searched articles of individual search terms.

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

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

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

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

Table 2. Scope of the selected sections.

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

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

3. Results

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

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

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

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

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

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

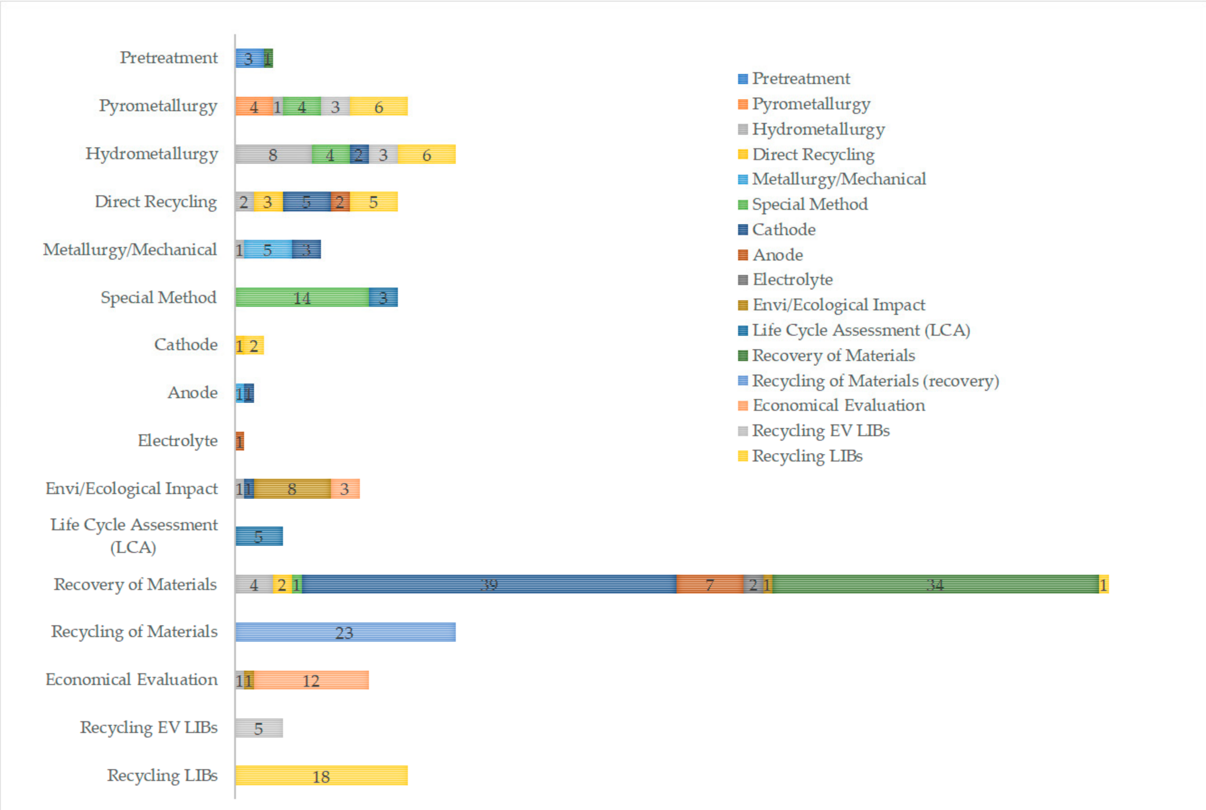

Figure 2. Total distribution and content overlap for publication selected according to the literature review in categories.

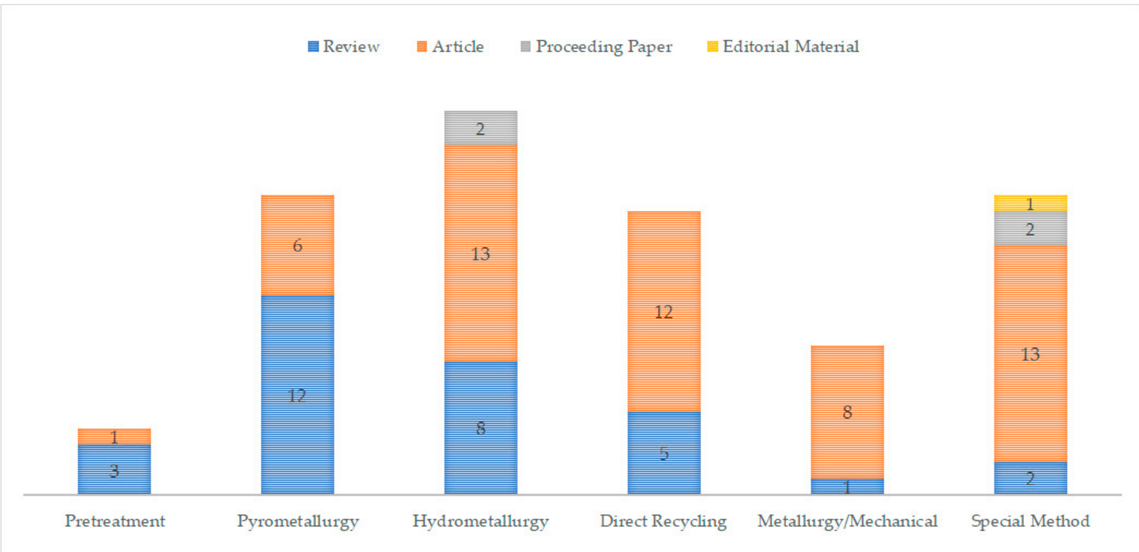

Figure 3. Distribution of "Recycling Technology" publications according to their type.

3.1. Pretreatment of the Recycling Processes

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

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

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

3.1.1. Laboratory-Scale Pretreatment Methods

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

1. Discharging

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

2. Dismantling

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

3. Separation

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

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

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

3.1.2. Industrial-Scale Pretreatment Methods

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

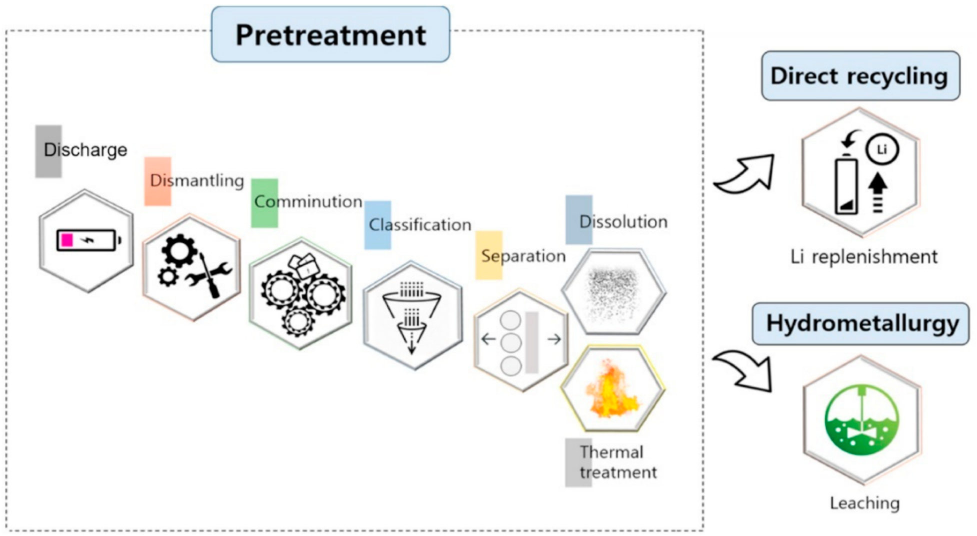

Figure 4. The scope and the sequence of the pretreatment process in LIB recycling [11]. Reprinted from Journal of Cleaner Production, Vol. 294, S. Kim et al., A comprehensive review on the pretreatment process in lithium-ion battery recycling, no. 8, pp. 1–29, Copyright (2021), with permission from Elsevier.

1. Discharging

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

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

2. Dismantling (disassembly)

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

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

3. Comminution (mechanical treatment)

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

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

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

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

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

4. Classification (sieving)

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

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

5. Separation

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

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

6. Dissolution

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

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

7. Thermal treatment

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

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

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

3.2. Recycling Processes

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

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

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

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

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

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

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

- means that information was not fund.

3.2.1. Metallurgical and Mechanical Processes

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

As outlined in the next sections, the recycling processes are created by long and complex process chains to accomplish the main recycling aims fulfilling the high environmental and economic requirements. In the past few years, various recycling procedures have been developed. These techniques can be divided into two general process routes: pretreatment of the processes (the mechanical treatment before the metallurgy) and a straight pyrometallurgy or hydrometallurgy technique, or their combination, e.g., the pyrometallurgy and subsequent hydrometallurgical treatment. Direct recycling is still in a phase of industrial implementation [10,21,33,38].

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

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

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

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

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

There are many approaches for the mechanical pretreatment of recycling LIBs. Thermal treatment can be applied before or after the mechanical methods or may be omitted altogether. The thermal treatment is typically formed by pyrolysis at around 500 °C to reduce the energy and eliminate the organic and halogenic content. The pyrolyzed LIBs can be safely mechanically processed without fire hazards [10,36]. In the other cases, specific safety measures are mandatory to prevent explosion and ignition during the processes. The non-thermal-treated procedures include for instance crushing under inert atmosphere of nitrogen (N_2), carbon dioxide (CO_2), or argon (Ar), followed by recovery of the volatile components by vacuum distillation or drying at moderate temperatures, or comminution in a solution, e.g., in a slightly alkaline medium [10,42].

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

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

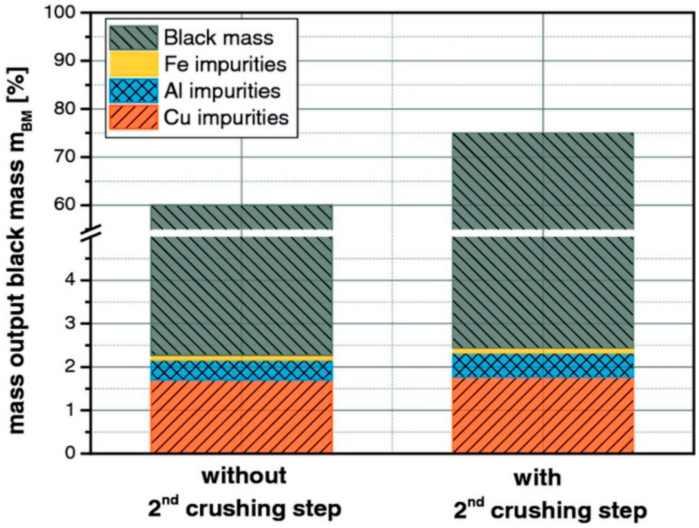

Figure 5. Yield of the black mass and impurities with/without 2nd crushing step [39]. Reprinted from Journal of the Electrochemical Society, Vol. 164, J. Diekmann et al., Ecological Recycling of Lithium-Ion Batteries from Electric Vehicles with Focus on Mechanical Processes, no. 1, pp. A6184–A6191, Copyright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

3.2.2. Pyrometallurgical Process

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

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

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

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

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

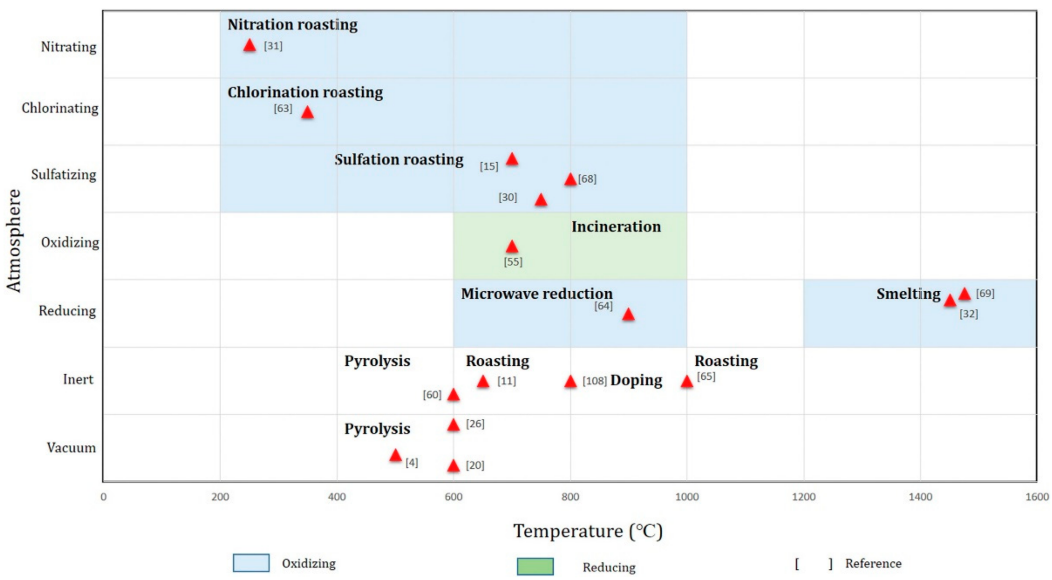

Figure 6. A brief data review of published work on pyrometallurgical options for recycling Li-ion batteries [43]. Reprinted from Journal of Power Sources, Vol. 491, B. Makuza et al., Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review, Copyright (2021), with permission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

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

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

The smelting process allows the recycling of various EOL LIBs based on different chemistries [9]. By smelting, the material of the retired battery is heated above its melting point and subsequently the metals are separate in the liquid phase by reduction and subsequent formation of immiscible molten layers [43,49]. Moreover, this process eliminates the necessity for a previous passivation step, and the battery cells can be directly thrown into the smelting furnace. The process of smelting usually consists of two complementary phases [43]:

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

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

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

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

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

3.2.3. Hydrometallurgical Process

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

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

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

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

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

peroxide (H_2O_2), and nitric acid (HNO_3) [59] or organic acids, e.g., citric acid, malic acid, oxalic acid, etc., are usually used [60,61].
- The second stage "Impurity removal" is created by solid-liquid separation, which clarifies the leach solution by filtration or centrifugation [55,62].
- The last stage "Ni, Co, Mn, and Li recovery" is devoted to the final recovery of valuable metals in hydroxide or metal salts [55]. This process includes, for example, solvent extraction [63], electrochemical techniques [64], selective precipitation [65], and separation by ion exchange resins [55].

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

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

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

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

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

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

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

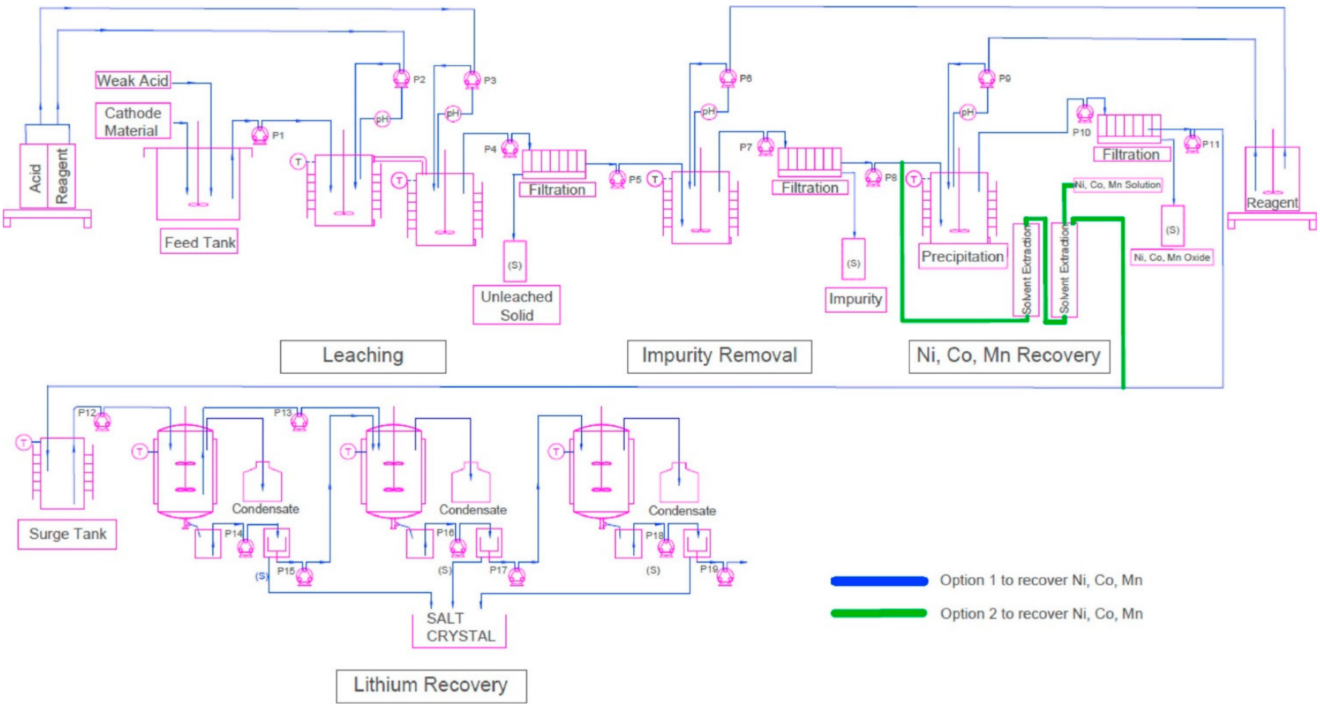

Figure 7. Schematic of the hydrometallurgy process for recycling battery cathode materials [58]. Reprinted from Journal of Energy Storage, Vol. 35, J. C. Y. Jung, P. C. Sui, and J. Zhang, A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments, Copyright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

3.2.4. Direct Recycling Process

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

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

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

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

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

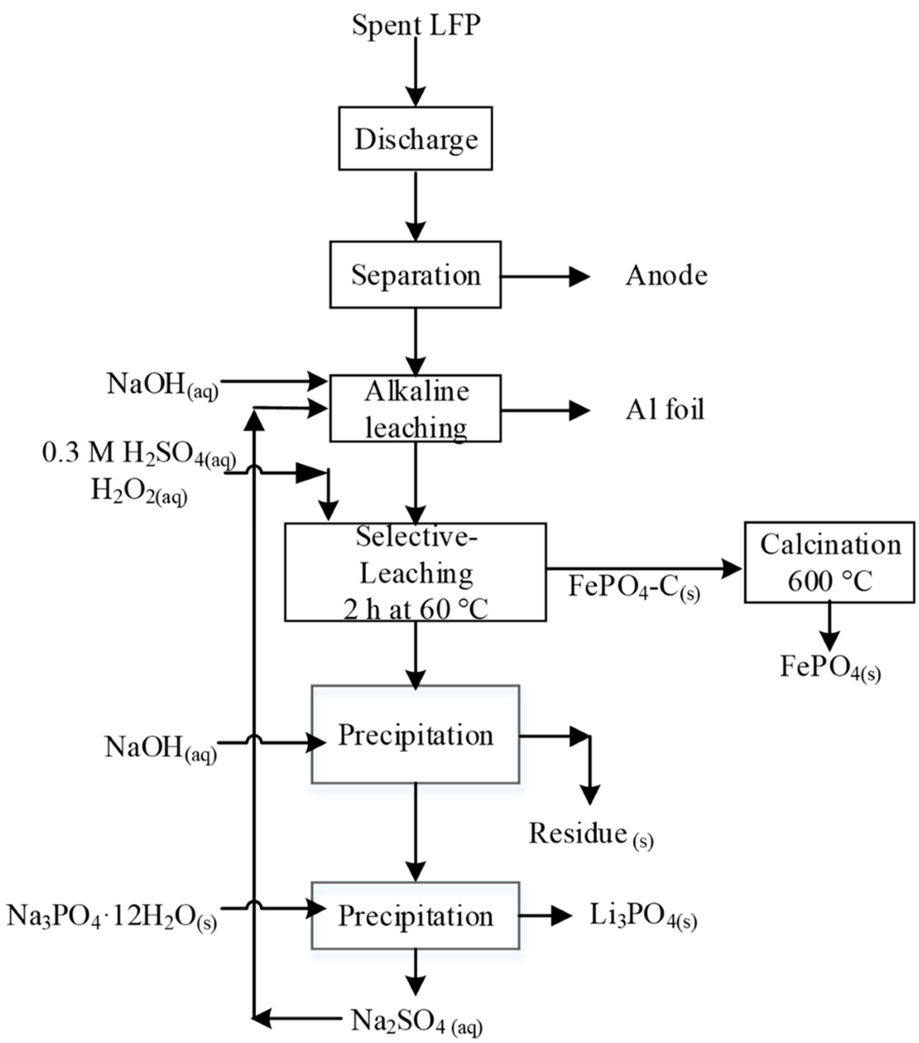

Figure 8. Selective leaching process [58]. Reprinted from Materials, Vol. 13, F. Larouche et al., Progress and Status of Hydrometallurgical and Direct Recycling of Li-Ion Batteries and Beyond, no. 3, p. 801, Copyright (2021), distributed under Creative Commons Attribution License.

Other direct recycling processes are being applied; for example, in a simple patented process by [79], very high-frequency ultrasound (>900 kHz) for up to 30 min was applied to the whole (not damaged) battery. Thereby the surface of the cathodes was cleaned out of absorbed phosphorous compounds that come from the dissociation of electrolyte lithium hexafluorophosphate (LiPF6) during cycling. Another leaching procedure was developed by [80], where the cathode materials are cleansed off residual Cu and Al in an alkaline solution (a complexing agent (5 M ammonium hydroxide NH_4OH) and 1 M lithium hydroxide (LiOH) with O_2 (g) bubbling to promote oxidation) after separation from other components. After 12 h, these contaminants are completely solubilized. Using the direct recycling procedure, any type of LIB can be treated. However, LFP batteries must comply with the specific pH requirements for process safety [58].

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

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

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

3.2.5. Special Recycling Methods

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

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

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

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

Existing industrial techniques are currently being extended under a laboratory scale. For example, using molten salts within a temperature range of 800–900 °C as electrolytes and reaction media is being applied as a chemical recycling procedure. Different eutectic mixtures of molten salts such as sodium, potassium, lithium, and calcium borates and chlorides, sodium, and potassium carbonates are being tested to provide an optimized alternative to the hydrometallurgical or pyrometallurgical processes of metal recovery. The final metal purity for single-metal oxides for these processes is around 98–99% [92].

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

4. Discussion and Conclusions

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

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

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

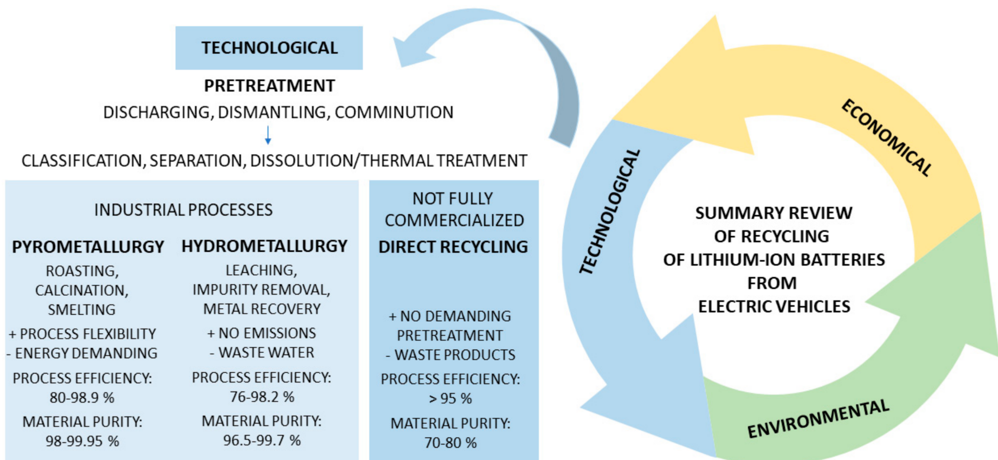

Figure 9. The summarizing figure of the first part of the literature review of recycling lithium-ion batteries from electric vehicles specialized on techniques of this process.

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

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

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

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

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

Although the overall efficiency of the mentioned processes is mostly above 90%, their implementation is limited by their technological complexity, required output products, further processing, predicted, or already monitored environmental burden, and costs that can be provided for line equipment and operation. Even though the batteries can be pyrometallurgical recycled on existing facilities, the operating costs of the whole process are high (mainly because the process requires high temperatures). In addition, toxic gases and water are released. Pyrometallurgy does not allow the recycling of electrolyte or graphite from anode active materials. On the contrary, the equipment of the hydrometallurgical line is costly, but the operating costs are much lower (depending on the chemicals used). As an intermediate product, only toxic wastewater is discharged and is further treated for reuse. Even hydrometallurgy does not allow the recycling of electrolytes from batteries [33,100,101].

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

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

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

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

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

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Acknowledgments: This review was discussed and supported by EEB/1 HV-Battery Systems and VAP eMobility Business Development Aftersales Škoda Auto, a.s.

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

Appendix A

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

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

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

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

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

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

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

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

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

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

Table A5. *Cont.*

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

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

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

References

1. Searchinger, T.D.; Hamburg, S.P.; Melillo, J.; Chameides, W.; Havlik, P.; Kammen, D.M.; Likens, G.E.; Lubowski, R.N.; Obersteiner, M.; Oppenheimer, M.; et al. Climate change. Fixing a critical climate accounting error. *Science* **2009**, *326*, 527–528. [CrossRef]
2. Lander, L.; Cleaver, T.; Rajaeifar, M.A.; Nguyen-Tien, V.; Elliott, R.J.R.; Heidrich, O.; Kendrick, E.; Edge, J.S.; Offer, G. Financial viability of electric vehicle lithium-ion battery recycling. *iScience* **2021**, *24*, 102787. [CrossRef] [PubMed]
3. Rahman, A.; Afroz, R.; Safrin, M. Recycling and disposal of lithium batteries: An economical and environmental approach. *IIUM Eng. J.* **2017**, *18*, 238–252. [CrossRef]
4. Jones, B.; Elliott, R.J.R.; Nguyen-Tien, V. The EV revolution: The road ahead for critical raw materials demand. *Appl. Energy* **2020**, *280*, 115072. [CrossRef] [PubMed]
5. Zheng, R.; Wang, W.; Dai, Y.; Ma, Q.; Liu, Y.; Mu, D.; Li, R.; Ren, J.; Dai, C. A closed-loop process for recycling LiNixCoyMn(1−x−y)O_2 from mixed cathode materials of lithium-ion batteries. *Green Energy Environ.* **2017**, *2*, 42–50. [CrossRef]
6. Kim, H.J.; Krishna, T.N.V.; Zeb, K.; Rajangam, V.; Muralee Gopi, C.V.V.; Sambasivam, S.; Raghavendra, K.V.G.; Obaidat, I.M. A comprehensive review of li-ion battery materials and their recycling techniques. *Electronics* **2020**, *9*, 1161. [CrossRef]
7. Assefi, M.; Maroufi, S.; Yamauchi, Y.; Sahajwalla, V. Pyrometallurgical recycling of Li-ion, Ni–Cd and Ni–MH batteries: A minireview. *Curr. Opin. Green Sustain. Chem.* **2020**, *24*, 26–31. [CrossRef]
8. Fan, E.; Li, L.; Wang, Z.; Lin, J.; Huang, Y.; Yao, Y.; Chen, R.; Wu, F. Sustainable Recycling Technology for Li-Ion Batteries and Beyond: Challenges and Future Prospects. *Chem. Rev.* **2020**, *120*, 7020–7063. [CrossRef]
9. Harper, G.; Sommerville, R.; Kendrick, E.; Driscoll, L.; Slater, P.; Stolkin, R.; Walton, A.; Christensen, P.; Heidrich, O.; Lambert, S.; et al. Recycling lithium-ion batteries from electric vehicles. *Nature* **2019**, *575*, 75–86. [CrossRef]
10. Brückner, L.; Frank, J.; Elwert, T. Industrial Recycling of Lithium-Ion Batteries—A Critical Review of Metallurgical Process Routes. *Metals* **2020**, *10*, 1107. [CrossRef]
11. Kim, S.; Bang, J.; Yoo, J.; Shin, Y.; Bae, J.; Jeong, J.; Kim, K.; Dong, P.; Kwon, K. A comprehensive review on the pretreatment process in lithium-ion battery recycling. *J. Clean. Prod.* **2021**, *294*, 126329. [CrossRef]
12. Zhang, G.; Yuan, X.; He, Y.; Wang, H.; Zhang, T.; Xie, W. Recent advances in pretreating technology for recycling valuable metals from spent lithium-ion batteries. *J. Hazard. Mater.* **2021**, *406*, 124332. [CrossRef] [PubMed]
13. Or, T.; Gourley, S.W.D.; Kaliyappan, K.; Yu, A.; Chen, Z. Recycling of mixed cathode lithium-ion batteries for electric vehicles: Current status and future outlook. *Carbon Energy* **2020**, *2*, 6–43. [CrossRef]
14. Yun, L.; Linh, D.; Shui, L.; Peng, X.; Garg, A.; LE, M.L.P.; Asghari, S.; Sandoval, J. Metallurgical and mechanical methods for recycling of lithium-ion battery pack for electric vehicles. *Resour. Conserv. Recycl.* **2018**, *136*, 198–208. [CrossRef]
15. Xiao, J.; Li, J.; Xu, Z. Recycling metals from lithium ion battery by mechanical separation and vacuum metallurgy. *J. Hazard. Mater.* **2017**, *338*, 124–131. [CrossRef]
16. Werner, D.; Peuker, U.A.; Mütze, T. Recycling chain for spent lithium-ion batteries. *Metals* **2020**, *10*, 316. [CrossRef]
17. Fujita, T.; Chen, H.; Wang, K.T.; He, C.L.; Wang, Y.B.; Dodbiba, G.; Wei, Y.-Z. Reduction, reuse and recycle of spent Li-ion batteries for automobiles: A review. *Int. J. Miner. Metall. Mater.* **2021**, *28*, 179–192. [CrossRef]
18. Pinegar, H.; Smith, Y.R. Recycling of End-of-Life Lithium Ion Batteries, Part I: Commercial Processes. *J. Sustain. Metall.* **2019**, *5*, 402–416. [CrossRef]
19. Park, K.; Yu, J.; Coyle, J.; Dai, Q.; Frisco, S.; Zhou, M.; Burrell, A. Direct Cathode Recycling of End-Of-Life Li-Ion Batteries Enabled by Redox Mediation. *ACS Sustain. Chem. Eng.* **2021**, *9*, 8214–8221. [CrossRef]
20. Chan, K.H.; Malik, M.; Anawati, J.; Azimi, G. Recycling of end-of-life lithium-ion battery of electric vehicles. In *Rare Metal Technology 2020*; Springer: Cham, Switzerland, 2020; pp. 23–32. [CrossRef]
21. Xu, J.; Thomas, H.R.; Francis, R.W.; Lum, K.R.; Wang, J.; Liang, B. A review of processes and technologies for the recycling of lithium-ion secondary batteries. *J. Power Sources* **2008**, *177*, 512–527. [CrossRef]
22. Vanitha, M.; Balasubramanian, N. Waste minimization and recovery of valuable metals from spent lithium-ion batteries—A review. *Environ. Technol. Rev.* **2013**, *2*, 101–115. [CrossRef]
23. Senćanski, J.; Bajuk-Bogdanović, D.; Majstorović, D.; Tchernychova, E.; Papan, J.; Vujković, M. The synthesis of Li(Co[sbnd]Mn[sbnd]Ni)O2 cathode material from spent-Li ion batteries and the proof of its functionality in aqueous lithium and sodium electrolytic solutions. *J. Power Sources* **2017**, *342*, 690–703. [CrossRef]
24. Sommerfeld, M.; Vonderstein, C.; Dertmann, C.; Klimko, J.; Oráč, D.; Miškufová, A.; Havlík, T.; Friedrich, B. A combined pyro-and hydrometallurgical approach to recycle pyrolyzed lithium-ion battery black mass part 1: Production of lithium concentrates in an electric arc furnace. *Metals* **2020**, *10*, 1069. [CrossRef]
25. Zhang, G.; He, Y.; Wang, H.; Feng, Y.; Xie, W.; Zhu, X. Removal of Organics by Pyrolysis for Enhancing Liberation and Flotation Behavior of Electrode Materials Derived from Spent Lithium-Ion Batteries. *ACS Sustain. Chem. Eng.* **2020**, *8*, 2205–2214. [CrossRef]
26. Vieceli, N.; Nogueira, C.A.; Guimarães, C.; Pereira, M.F.C.; Durão, F.O.; Margarido, F. Hydrometallurgical recycling of lithium-ion batteries by reductive leaching with sodium metabisulphite. *Waste Manag.* **2018**, *71*, 350–361. [CrossRef] [PubMed]
27. Shin, S.M.; Kim, N.H.; Sohn, J.S.; Yang, D.H.; Kim, Y.H. Development of a metal recovery process from Li-ion battery wastes. *Hydrometallurgy* **2005**, *79*, 172–181. [CrossRef]
28. Bi, H.; Zhu, H.; Zu, L.; Bai, Y.; Gao, S.; Gao, Y. A new model of trajectory in eddy current separation for recovering spent lithium iron phosphate batteries. *Waste Manag.* **2019**, *100*, 1–9. [CrossRef] [PubMed]

29. Silveira, A.V.M.; Santana, M.P.; Tanabe, E.H.; Bertuol, D.A. Recovery of valuable materials from spent lithium ion batteries using electrostatic separation. *Int. J. Miner. Process.* **2017**, *169*, 91–98. [CrossRef]
30. Meng, Q.; Zhang, Y.; Dong, P. Use of glucose as reductant to recover Co from spent lithium ions batteries. *Waste Manag.* **2017**, *64*, 214–218. [CrossRef]
31. Liu, Y.J.; Hu, Q.Y.; Li, X.H.; Wang, Z.X.; Guo, H.J. Recycle and synthesis of $LiCoO_2$ from incisors bound of Li-ion batteries. *Trans. Nonferrous Met. Soc. China Engl. Ed.* **2006**, *16*, 956–959. [CrossRef]
32. Sun, L.; Qiu, K. Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries. *J. Hazard. Mater.* **2011**, *194*, 378–384. [CrossRef] [PubMed]
33. Northic Council of Ministers. *Mapping of Lithium-Ion Batteries for Vehicles*; Nordisk Ministerråd: Copenhagen, Denmark, 2019; ISBN 9789289362931. [CrossRef]
34. Chen, M.; Ma, X.; Chen, B.; Arsenault, R.; Karlson, P.; Simon, N.; Wang, Y. Recycling End-of-Life Electric Vehicle Lithium-Ion Batteries. *Joule* **2019**, *3*, 2622–2646. [CrossRef]
35. Rallo Tolós, H. Second Life Batteries of Electric Vehicles: Analysis of Use and Management Models. 2021. Available online: https://upcommons.upc.edu/handle/2117/346658 (accessed on 16 September 2021).
36. Georgi-Maschler, T.; Friedrich, B.; Weyhe, R.; Heegn, H.; Rutz, M. Development of a recycling process for Li-ion batteries. *J. Power Sources* **2012**, *207*, 173–182. [CrossRef]
37. Zenger Method of and Apparatus for Dismantling and Storage of Objects Comprising Alkali Metals, Such as Alkali Metal Containing Batteries. IFI CLAIMS Patent Services: New Haven, USA. 2003. Available online: https://patents.google.com/patent/US7833646B2/en (accessed on 18 December 2021).
38. Yao, Y.; Zhu, M.; Zhao, Z.; Tong, B.; Fan, Y.; Hua, Z. Hydrometallurgical Processes for Recycling Spent Lithium-Ion Batteries: A Critical Review. *ACS Sustain. Chem. Eng.* **2018**, *6*, 13611–13627. [CrossRef]
39. Diekmann, J.; Hanisch, C.; Froböse, L.; Schälicke, G.; Loellhoeffel, T.; Fölster, A.-S.; Kwade, A. Ecological Recycling of Lithium-Ion Batteries from Electric Vehicles with Focus on Mechanical Processes. *J. Electrochem. Soc.* **2017**, *164*, A6184–A6191. [CrossRef]
40. Li, L.; Zhang, X.; Li, M.; Chen, R.; Wu, F.; Amine, K.; Lu, J. The Recycling of Spent Lithium-Ion Batteries: A Review of Current Processes and Technologies. *Electrochem. Energy Rev.* **2018**, *1*, 461–482. [CrossRef]
41. Mayyas, A.; Steward, D.; Mann, M. The Case for Recycling: Overview and Challenges in the Material Supply Chain for Automotive Li-Ion Batteries. *Sustain. Mater. Technol.* **2018**, *19*, e00087. [CrossRef]
42. Hanisch, C. Recycling Method for Treating Used Batteries, In Particular Rechergeable Batteries, and Battery Processing Installation. Canada Patent EP3312922B1, 15 August 2018.
43. Makuza, B.; Tian, Q.; Guo, X.; Chattopadhyay, K.; Yu, D. Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. *J. Power Sources* **2021**, *491*, 229622. [CrossRef]
44. Cerrillo-Gonzalez, M.d.M.; Villen-Guzman, M.; Acedo-Bueno, L.F.; Rodriguez-Maroto, J.M.; Paz-Garcia, J.M. Hydrometallurgical extraction of Li and Co from $LiCoO_2$ particles-experimental and modeling. *Appl. Sci.* **2020**, *10*, 6375. [CrossRef]
45. Haldar, S.K. Mineral Processing. *Miner. Explor.* **2018**, 259–290. [CrossRef]
46. Barker, J.; Saidi, M.Y.; Swoyer, J.L. Lithium iron(II) phospho-olivines prepared by a novel carbothermal reduction method. *Electrochem. Solid-State Lett.* **2003**, *6*, A53. [CrossRef]
47. Li, J.; Wang, G.; Xu, Z. Environmentally-friendly oxygen-free roasting/wet magnetic separation technology for in situ recycling cobalt, lithium carbonate and graphite from spent $LiCoO_2$/graphite lithium batteries. *J. Hazard. Mater.* **2016**, *302*, 97–104. [CrossRef] [PubMed]
48. Zhang, X.; Li, L.; Fan, E.; Xue, Q.; Bian, Y.; Wu, F.; Chen, R. Toward sustainable and systematic recycling of spent rechargeable batteries. *Chem. Soc. Rev.* **2018**, *47*, 7239–7302. [CrossRef]
49. Sohn, H.Y.; Wadsworth, M.E. *Rate Processes of Extractive Metallurgy*; Springer: New York, NY, USA, 1979. [CrossRef]
50. Gaines, L.; Richa, K.; Spangenberger, J. Key issues for Li-ion battery recycling. *MRS Energy Sustain.* **2018**, *5*, 12. [CrossRef]
51. Beaudet, A.; Larouche, F.; Amouzegar, K.; Bouchard, P.; Zaghib, K. Key Challenges and Opportunities for Recycling Electric Vehicle Battery Materials. *Sustainability* **2020**, *12*, 5837. [CrossRef]
52. Jung, J.C.Y.; Sui, P.C.; Zhang, J. A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments. *J. Energy Storage* **2021**, *35*, 102217. [CrossRef]
53. Chen, X.; Chen, Y.; Zhou, T.; Liu, D.; Hu, H.; Fan, S. Hydrometallurgical recovery of metal values from sulfuric acid leaching liquor of spent lithium-ion batteries. *Waste Manag.* **2015**, *38*, 349–356. [CrossRef]
54. Cerrillo-Gonzalez, M.M.; Villen-Guzman, M.; Vereda-Alonso, C.; Gomez-Lahoz, C.; Rodriguez-Maroto, J.M.; Paz-Garcia, J.M. Recovery of Li and Co from $LiCoO_2$ via hydrometallurgical-electrodialytic treatment. *Appl. Sci.* **2020**, *10*, 2367. [CrossRef]
55. Djoudi, N.; Le Page Mostefa, M.; Muhr, H. Hydrometallurgical process to recover cobalt from spent li-ion batteries. *Resources* **2021**, *10*, 58. [CrossRef]
56. He, Y.; Yuan, X.; Zhang, G.; Wang, H.; Zhang, T.; Xie, W.; Li, L. A critical review of current technologies for the liberation of electrode materials from foils in the recycling process of spent lithium-ion batteries. *Sci. Total Environ.* **2021**, *766*, 142382. [CrossRef]
57. Zhou, L.F.; Yang, D.; Du, T.; Gong, H.; Luo, W. Bin The Current Process for the Recycling of Spent Lithium Ion Batteries. *Front. Chem.* **2020**, *8*, 578044. [CrossRef] [PubMed]

58. Larouche, F.; Tedjar, F.; Amouzegar, K.; Houlachi, G.; Bouchard, P.; Demopoulos, G.P.; Zaghib, K. Progress and status of hydrometallurgical and direct recycling of Li-Ion batteries and beyond. *Materials* **2020**, *13*, 801. [CrossRef] [PubMed]
59. Meshram, P.; Abhilash; Pandey, B.D.; Mankhand, T.R.; Deveci, H. Comparision of Different Reductants in Leaching of Spent Lithium Ion Batteries. *JOM* **2016**, *68*, 2613–2623. [CrossRef]
60. Chen, X.; Zhou, T. Hydrometallurgical process for the recovery of metal values from spent lithium-ion batteries in citric acid media. *Waste Manag. Res.* **2014**, *32*, 1083–1093. [CrossRef] [PubMed]
61. Li, L.; Bian, Y.; Zhang, X.; Guan, Y.; Fan, E.; Wu, F.; Chen, R. Process for recycling mixed-cathode materials from spent lithium-ion batteries and kinetics of leaching. *Waste Manag.* **2018**, *71*, 362–371. [CrossRef] [PubMed]
62. Chagnes, A.; Pospiech, B. A brief review on hydrometallurgical technologies for recycling spent lithium-ion batteries. *J. Chem. Technol. Biotechnol.* **2013**, *88*, 1191–1199. [CrossRef]
63. Chen, L.; Tang, X.; Zhang, Y.; Li, L.; Zeng, Z.; Zhang, Y. Process for the recovery of cobalt oxalate from spent lithium-ion batteries. *Hydrometallurgy* **2011**, *108*, 80–86. [CrossRef]
64. Lupi, C.; Pasquali, M.; Dell'Era, A. Nickel and cobalt recycling from lithium-ion batteries by electrochemical processes. *Waste Manag.* **2005**, *25*, 215–220. [CrossRef] [PubMed]
65. Prabaharan, G.; Barik, S.P.; Kumar, N.; Kumar, L. Electrochemical process for electrode material of spent lithium ion batteries. *Waste Manag.* **2017**, *68*, 527–533. [CrossRef] [PubMed]
66. Zhang, P.; Yokoyama, T.; Itabashi, O.; Suzuki, T.M.; Inoue, K. Hydrometallurgical process for recovery of metal values from spent lithium-ion secondary batteries. *Hydrometallurgy* **1998**, *47*, 259–271. [CrossRef]
67. Contestabile, M.; Panero, S.; Scrosati, B. A laboratory-scale lithium-ion battery recycling process. *J. Power Sources* **2001**, *92*, 65–69. [CrossRef]
68. Takacova, Z.; Havlik, T.; Kukurugya, F.; Orac, D. Cobalt and lithium recovery from active mass of spent Li-ion batteries: Theoretical and experimental approach. *Hydrometallurgy* **2016**, *163*, 9–17. [CrossRef]
69. Wang, R.C.; Lin, Y.C.; Wu, S.H. A novel recovery process of metal values from the cathode active materials of the lithium-ion secondary batteries. *Hydrometallurgy* **2009**, *99*, 194–201. [CrossRef]
70. Castillo, S.; Ansart, F.; Laberty-Robert, C.; Portal, J. Advances in the recovering of spent lithium battery compounds. *J. Power Sources* **2002**, *112*, 247–254. [CrossRef]
71. Lee, C.K.; Rhee, K.I. Preparation of $LiCoO_2$ from spent lithium-ion batteries. *J. Power Sources* **2002**, *109*, 17–21. [CrossRef]
72. Arshad, F.; Li, L.; Amin, K.; Fan, E.; Manurkar, N.; Ahmad, A.; Yang, J.; Wu, F.; Chen, R. A Comprehensive Review of the Advancement in Recycling the Anode and Electrolyte from Spent Lithium Ion Batteries. *ACS Sustain. Chem. Eng.* **2020**, *8*, 13527–13554. [CrossRef]
73. Sloop, S.; Crandon, L.; Allen, M.; Koetje, K.; Reed, L.; Gaines, L.; Sirisaksoontorn, W.; Lerner, M. A direct recycling case study from a lithium-ion battery recall. *Sustain. Mater. Technol.* **2020**, *25*, e00152. [CrossRef]
74. Li, L.; Yang, T.; Li, Z. Parameter optimization and yield prediction of cathode coating separation process for direct recycling of end-of-life lithium-ion batteries. *RSC Adv.* **2021**, *11*, 24132–24136. [CrossRef]
75. Smith, W.N.; Swoffer, S. Smith Process for Recovering and Regenerating Lithium Cathode Material from Lithium-Ion Batteries. U.S. Patent US8882007B1, 21 November 2013.
76. Kim, H.S.; Shin, E.J. Re-synthesis and Electrochemical Characteristics of $LiFePO_4$ Cathode Materials Recycled from Scrap Electrodes. *Bull. Korean Chem. Soc.* **2013**, *34*, 851–855. [CrossRef]
77. Chen, J.; Li, Q.; Song, J.; Song, D.; Zhang, L.; Shi, X. Environmentally friendly recycling and effective repairing of cathode powders from spent $LiFePO_4$ batteries. *Green Chem.* **2016**, *18*, 2500–2506. [CrossRef]
78. Wang, L.; Li, J.; Zhou, H.; Huang, Z.; Tao, S.; Zhai, B.; Liu, L.; Hu, L. Regeneration cathode material mixture from spent lithium iron phosphate batteries. *J. Mater. Sci. Mater. Electron.* **2018**, *29*, 9283–9290. [CrossRef]
79. Okayama, S.; Uchida, S. Degraded Performance Recovery Method for Lithium Ion Secondary Battery. U.S. Patent US9958508B2, 1 May 2018.
80. Tsang, F.; Hailey, P. Method for Removing Copper and Aluminum from an Electrode Material, and Process for Recycling Electrode Material from Waste Lithium-Ion Batteries. U.S. Patent US10103413B2, 16 October 2018.
81. Mohr, M.; Peters, J.F.; Baumann, M.; Weil, M. Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes. *J. Ind. Ecol.* **2020**, *24*, 1310–1322. [CrossRef]
82. Tang, Y.; Zhang, Q.; Li, Y.; Wang, G.; Li, Y. Recycling mechanisms and policy suggestions for spent electric vehicles' power battery -A case of Beijing. *J. Clean. Prod.* **2018**, *186*, 388–406. [CrossRef]
83. Silvestri, L.; Forcina, A.; Arcese, G.; Bella, G. Recycling technologies of nickel–metal hydride batteries: An LCA based analysis. *J. Clean. Prod.* **2020**, *273*, 123083. [CrossRef]
84. Gaines, L.; Sullivan, J.; Burnham, A.; Belharouak, I. Life-cycle analysis of production and recycling of lithium ion batteries. *Transp. Res. Rec.* **2011**, 57–65. [CrossRef]
85. Sommerville, R.; Shaw-Stewart, J.; Goodship, V.; Rowson, N.; Kendrick, E. A review of physical processes used in the safe recycling of lithium ion batteries. *Sustain. Mater. Technol.* **2020**, *25*, e00197. [CrossRef]
86. Gaines, L. Lithium-ion battery recycling processes: Research towards a sustainable course. *Sustain. Mater. Technol.* **2018**, *17*, e00068. [CrossRef]

87. Diekmann, J.; Hanisch, C.; Loellhoeffel, T.; Schalicke, G.; Kwade, A. (Invited) Ecologically Friendly Recycling of Lithium-Ion Batteries—The LithoRec Process. *ECS Trans.* **2016**, *73*, 1–9. [CrossRef]
88. Norgren, A.; Carpenter, A.; Heath, G. Design for Recycling Principles Applicable to Selected Clean Energy Technologies: Crystalline-Silicon Photovoltaic Modules, Electric Vehicle Batteries, and Wind Turbine Blades. *J. Sustain. Metall.* **2020**, *6*, 761–774. [CrossRef]
89. Zhang, T.; He, Y.; Ge, L.; Fu, R.; Zhang, X.; Huang, Y. Characteristics of wet and dry crushing methods in the recycling process of spent lithium-ion batteries. *J. Power Sources* **2013**, *240*, 766–771. [CrossRef]
90. Gaines, L. Profitable Recycling of Low-Cobalt Lithium-Ion Batteries Will Depend on New Process Developments. *One Earth* **2019**, *1*, 413–415. [CrossRef]
91. Li, Y.; Mao, J.; Xie, H.; Li, J. Heat-treatment recycling of waste toner and its applications in lithium ion batteries. *J. Mater. Cycles Waste Manag.* **2018**, *20*, 361–368. [CrossRef]
92. Carvajal-Ortiz, R. Alternative recycling process for lithium-ion batteries: Molten salt approach. *Johnson Matthey Technol. Rev.* **2020**, *64*, 16–18. [CrossRef]
93. Wang, M.M.; Zhang, C.C.; Zhang, F.S. Recycling of spent lithium-ion battery with polyvinyl chloride by mechanochemical process. *Waste Manag.* **2017**, *67*, 232–239. [CrossRef] [PubMed]
94. Cognet, M.; Condomines, J.; Cambedouzou, J.; Madhavi, S.; Carboni, M.; Meyer, D. An original recycling method for Li-ion batteries through large scale production of Metal Organic Frameworks. *J. Hazard. Mater.* **2020**, *385*, 121603. [CrossRef]
95. Pindar, S.; Dhawan, N. Recycling of mixed discarded lithium-ion batteries via microwave processing route. *Sustain. Mater. Technol.* **2020**, *25*, e00157. [CrossRef]
96. Kotak, Y.; Fernández, C.M.; Casals, L.C.; Kotak, B.S.; Koch, D.; Geisbauer, C.; Trilla, L.; Gómez-Núñez, A.; Schweiger, H.G. End of Electric Vehicle Batteries: Reuse vs. Recycle. *Energies* **2021**, *14*, 2217. [CrossRef]
97. Franco, M.A.; Groesser, S.N. A Systematic Literature Review of the Solar Photovoltaic Value Chain for a Circular Economy. *Sustainability* **2021**, *13*, 9615. [CrossRef]
98. Bae, H.; Kim, Y. Technologies of lithium recycling from waste lithium ion batteries: A review. *Mater. Adv.* **2021**, *2*, 3234–3250. [CrossRef]
99. Holzer, A.; Windisch-Kern, S.; Ponak, C.; Raupenstrauch, H. A novel pyrometallurgical recycling process for lithium-ion batteries and its application to the recycling of lco and lfp. *Metals* **2021**, *11*, 149. [CrossRef]
100. Chitre, A.; Freake, D.; Lander, L.; Edge, J.; Titirici, M. Towards a More Sustainable Lithium-Ion Battery Future: Recycling LIBs from Electric Vehicles. *Batter. Supercaps* **2020**, *3*, 1126–1136. [CrossRef]
101. Thomas, M.; Ager-Wick Ellingsen, L.; Roxanne Hung, C. Research for TRAN Committee-Battery-Powered Electric Vehicles: Market Development and Lifecycle Emissions. 2018. Available online: https://www.europarl.europa.eu/thinktank/en/document/IPOL_STU(2018)617457 (accessed on 18 December 2021).
102. Meng, F.; McNeice, J.; Zadeh, S.S.; Ghahreman, A. Review of Lithium Production and Recovery from Minerals, Brines, and Lithium-Ion Batteries. *Miner. Process. Extr. Metall. Rev.* **2021**, *42*, 123–141. [CrossRef]
103. Lombardo, G.; Ebin, B.; Mark, M.R.; Steenari, B.M.; Petranikova, M. Incineration of EV Lithium-ion batteries as a pretreatment for recycling—Determination of the potential formation of hazardous by-products and effects on metal compounds. *J. Hazard. Mater.* **2020**, *393*, 122372. [CrossRef] [PubMed]
104. Xiao, J.; Li, J.; Xu, Z. Novel Approach for in Situ Recovery of Lithium Carbonate from Spent Lithium Ion Batteries Using Vacuum Metallurgy. *Environ. Sci. Technol.* **2017**, *51*, 11960–11966. [CrossRef]
105. Porvali, A.; Aaltonen, M.; Ojanen, S.; Velazquez-Martinez, O.; Eronen, E.; Liu, F.; Wilson, B.P.; Serna-Guerrero, R.; Lundström, M. Mechanical and hydrometallurgical processes in HCl media for the recycling of valuable metals from Li-ion battery waste. *Resour. Conserv. Recycl.* **2019**, *142*, 257–266. [CrossRef]
106. Munir, H.; Srivastava, R.R.; Kim, H.; Ilyas, S.; Khosa, M.K.; Yameen, B. Leaching of exhausted LNCM cathode batteries in ascorbic acid lixiviant: A green recycling approach, reaction kinetics and process mechanism. *J. Chem. Technol. Biotechnol.* **2020**, *95*, 2286–2294. [CrossRef]
107. Murakami, Y.; Matsuzaki, Y.; Kamimura, T.; Nishiura, T.; Masuda, K.; Shibayama, A.; Inoue, R. Erosion mechanism of refractories in a pyro-processing furnace for recycling lithium-ion secondary batteries. *Ceram. Int.* **2020**, *46*, 9281–9288. [CrossRef]
108. Kader, Z.A.; Marshall, A.; Kennedy, J. A review on sustainable recycling technologies for lithium-ion batteries. *Emergent Mater.* **2021**, *4*, 725–735. [CrossRef]
109. Hu, Y.; Yu, Y.; Huang, K.; Wang, L. Development tendency and future response about the recycling methods of spent lithium-ion batteries based on bibliometrics analysis. *J. Energy Storage* **2020**, *27*, 101111. [CrossRef]
110. Heelan, J.; Gratz, E.; Zheng, Z.; Wang, Q.; Chen, M.; Apelian, D.; Wang, Y. Current and Prospective Li-Ion Battery Recycling and Recovery Processes. *JOM* **2016**, *68*, 2632–2638. [CrossRef]
111. Zeng, X.; Li, J.; Singh, N. Recycling of spent lithium-ion battery: A critical review. *Crit. Rev. Environ. Sci. Technol.* **2014**, *44*, 1129–1165. [CrossRef]
112. Atia, T.A.; Elia, G.; Hahn, R.; Altimari, P.; Pagnanelli, F. Closed-loop hydrometallurgical treatment of end-of-life lithium ion batteries: Towards zero-waste process and metal recycling in advanced batteries. *J. Energy Chem.* **2019**, *35*, 220–227. [CrossRef]
113. Ferreira, D.A.; Prados, L.M.Z.; Majuste, D.; Mansur, M.B. Hydrometallurgical separation of aluminium, cobalt, copper and lithium from spent Li-ion batteries. *J. Power Sources* **2009**, *187*, 238–246. [CrossRef]

114. Vieceli, N.; Casasola, R.; Lombardo, G.; Ebin, B.; Petranikova, M. Hydrometallurgical recycling of EV lithium-ion batteries: Effects of incineration on the leaching efficiency of metals using sulfuric acid. *Waste Manag.* **2021**, *125*, 192–203. [CrossRef]
115. Wang, H.; Friedrich, B. Development of a Highly Efficient Hydrometallurgical Recycling Process for Automotive Li–Ion Batteries. *J. Sustain. Metall.* **2015**, *1*, 168–178. [CrossRef]
116. Ciez, R.E.; Whitacre, J.F. Examining different recycling processes for lithium-ion batteries. *Nat. Sustain.* **2019**, *2*, 148–156. [CrossRef]
117. Wang, W.; Wu, Y. An overview of recycling and treatment of spent $LiFePO_4$ batteries in China. *Resour. Conserv. Recycl.* **2017**, *127*, 233–243. [CrossRef]
118. Ku, H.; Jung, Y.; Jo, M.; Park, S.; Kim, S.; Yang, D.; Rhee, K.; An, E.M.; Sohn, J.; Kwon, K. Recycling of spent lithium-ion battery cathode materials by ammoniacal leaching. *J. Hazard. Mater.* **2016**, *313*, 138–146. [CrossRef]
119. Sonoc, A.; Jeswiet, J. A review of lithium supply and demand and a preliminary investigation of a room temperature method to recycle lithium ion batteries to recover lithium and other materials. *Procedia CIRP* **2014**, *15*, 289–293. [CrossRef]
120. Wang, Y.; An, N.; Wen, L.; Wang, L.; Jiang, X.; Hou, F.; Yin, Y.; Liang, J. Recent progress on the recycling technology of Li-ion batteries. *J. Energy Chem.* **2020**, *55*, 391–419. [CrossRef]
121. Cai, G.; Fung, K.Y.; Ng, K.M.; Wibowo, C. Process development for the recycle of spent lithium ion batteries by chemical precipitation. *Ind. Eng. Chem. Res.* **2014**, *53*, 18245–18259. [CrossRef]
122. Ross, B.J.; Leresche, M.; Liu, D.; Durham, J.L.; Dahl, E.U.; Lipson, A.L. Mitigating the Impact of Thermal Binder Removal for Direct Li-Ion Battery Recycling. *ACS Sustain. Chem. Eng.* **2020**, *8*, 12511–12515. [CrossRef]
123. Fink, K.; Santhanagopalan, S.; Hartig, J.; Cao, L. Characterization of Aged Li-Ion Battery Components for Direct Recycling Process Design. *J. Electrochem. Soc.* **2019**, *166*, A3775–A3783. [CrossRef]
124. Moradi, B.; Botte, G.G. Recycling of graphite anodes for the next generation of lithium ion batteries. *J. Appl. Electrochem.* **2016**, *46*, 123–148. [CrossRef]
125. Hou, H.; Dai, Z.; Liu, X.; Yao, Y.; Yu, C.; Li, D. Direct and indirect recycling strategies of expired oxytetracycline for the anode material in lithium ion batteries. *Front. Mater.* **2019**, *6*, 80. [CrossRef]
126. Xu, P.; Dai, Q.; Gao, H.; Liu, H.; Zhang, M.; Li, M.; Chen, Y.; An, K.; Meng, Y.S.; Liu, P.; et al. Efficient Direct Recycling of Lithium-Ion Battery Cathodes by Targeted Healing. *Joule* **2020**, *4*, 2609–2626. [CrossRef]
127. Wang, H.; Whitacre, J.F. Direct Recycling of Aged $LiMn_2O_4$ Cathode Materials used in Aqueous Lithium-ion Batteries: Processes and Sensitivities. *Energy Technol.* **2018**, *6*, 2429–2437. [CrossRef]
128. Zheng, Z.; Chen, M.; Wang, Q.; Zhang, Y.; Ma, X.; Shen, C.; Xu, D.; Liu, J.; Liu, Y.; Gionet, P.; et al. High Performance Cathode Recovery from Different Electric Vehicle Recycling Streams. *ACS Sustain. Chem. Eng.* **2018**, *6*, 13977–13982. [CrossRef]
129. Gratz, E.; Sa, Q.; Apelian, D.; Wang, Y. A closed loop process for recycling spent lithium ion batteries. *J. Power Sources* **2014**, *262*, 255–262. [CrossRef]
130. Lv, W.; Wang, Z.; Cao, H.; Sun, Y.; Zhang, Y.; Sun, Z. A Critical Review and Analysis on the Recycling of Spent Lithium-Ion Batteries. *ACS Sustain. Chem. Eng.* **2018**, *6*, 1504–1521. [CrossRef]
131. Garole, D.J.; Hossain, R.; Garole, V.J.; Sahajwalla, V.; Nerkar, J.; Dubal, D.P. Recycle, Recover and Repurpose Strategy of Spent Li-ion Batteries and Catalysts: Current Status and Future Opportunities. *ChemSusChem* **2020**, *13*, 3079–3100. [CrossRef] [PubMed]
132. Sommerville, R.; Zhu, P.; Rajaeifar, M.A.; Heidrich, O.; Goodship, V.; Kendrick, E. A qualitative assessment of lithium ion battery recycling processes. *Resour. Conserv. Recycl.* **2021**, *165*, 105219. [CrossRef]
133. Lv, W.; Wang, Z.; Cao, H.; Zheng, X.; Jin, W.; Zhang, Y.; Sun, Z. A sustainable process for metal recycling from spent lithium-ion batteries using ammonium chloride. *Waste Manag.* **2018**, *79*, 545–553. [CrossRef] [PubMed]
134. Chen, L.; Wang, P.; Shen, Y.; Guo, M. Spent lithium-ion battery materials recycling for catalytic pyrolysis or gasification of biomass. *Bioresour. Technol.* **2021**, *323*, 124584. [CrossRef] [PubMed]
135. Bokelmann, K.; Horn, D.; Zimmermann, J.; Gellermann, C.; Stauber, R. Recycling von Li-Ionen-Batterien: Elektrohydraulische Zerkleinerung. *Chem. Unserer Zeit* **2018**, *52*, 284–285. [CrossRef]