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Review

A Review of Lithium-Ion Battery Recycling: Technologies, Sustainability, and Open Issues

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Abstract: Lithium-ion batteries (LIBs) are a widely used energy storage technology as they possess high energy density and are characterized by the reversible intercalation/deintercalation of Li ions between electrodes. The rapid development of LIBs has led to increased production efficiency and lower costs for manufacturers, resulting in a growing demand for batteries and their application across various industries, particularly in different types of vehicles. In order to meet the demand for LIBs while minimizing climate-impacting emissions, the reuse, recycling, and repurposing of LIBs is a critical step toward achieving a sustainable battery economy. This paper provides a comprehensive review of lithium-ion battery recycling, covering topics such as current recycling technologies, technological advancements, policy gaps, design strategies, funding for pilot projects, and a comprehensive strategy for battery recycling. Additionally, this paper emphasizes the challenges associated with developing LIB recycling and the opportunities arising from these challenges, such as the potential for innovation and the creation of a more sustainable and circular economy. The environmental implications of LIB recycling are also evaluated with methodologies able to provide a sustainability analysis of the selected technology. This paper aims to enhance the comprehension of these trade-offs and encourage discussion on determining the "best" recycling route when targets are in conflict.

Keywords: battery recycling; hydrometallurgy; pyrometallurgy; waste battery management; sustainability analysis (ESCAPE method)

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1. Introduction

Lithium-ion batteries (LIBs) have become increasingly significant as an energy storage technology since their introduction to the market in the early 1990s, owing to their high energy density [1]. Today, LIB technology is based on the so-called "intercalation chemistry", the key to their success, with both the cathode and anode materials characterized by a peculiar structure allowing for the reversible intercalation/deintercalation of Li ions shuffling between the electrodes [2]. As a result, portable electronics like mobile phones, laptops, and tablets have undergone rapid development. Additionally, in the 2010s, the usage of lithium-ion technology extended to electric and hybrid cars, buses, and energy storage systems.

LIBs work through a topochemical cell reaction, where lithium ions migrate between positive and negative electrodes. This migration of lithium ions allows for the storage and release of energy within the battery. LIBs consist of several key components, including the cathode, anode, electrolyte, and separator (see Figure 1). The cathode is typically made of layered oxide materials, such as $LiCoO_2$, which undergo reversible intercalation and deintercalation of sodium ions during charge and discharge processes. The anode is usually made of graphite or other carbon-based materials, which can intercalate lithium ions during charging [3]. The electrolyte, which is typically a lithium salt dissolved in an organic solvent, facilitates the movement of lithium ions between the electrodes. The separator, a key component for high-temperature LIBs, provides thermal stability, mechanical strength, and ionic conductivity. During operation, lithium ions flow from the anode to the cathode through the electrolyte, while electrons flow through an external circuit, creating a flow of electrical current. This process is reversible, allowing for the repeated charging and discharging of the battery [4].

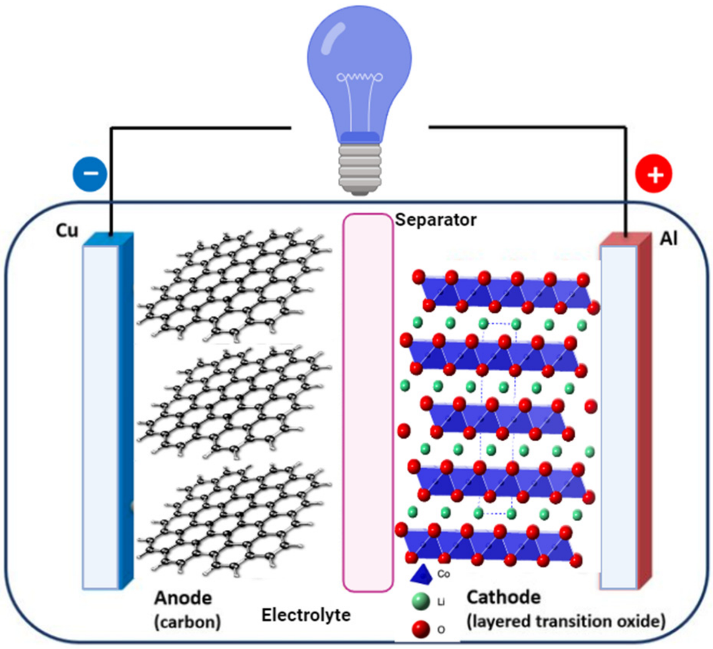


Figure 1. Key components of LIBs: cathode, anode, electrolyte, and separator.

Some significant milestones in the history of lithium-ion batteries and the expected future perspectives are reported in Figure 2. The first idea of intercalation chemistry was developed independently by Goodenough, Whittingham, and Yoshino in the 1980s and led to the development of the first LIB prototype based on a $LiCoO_2$ layered structure as the cathode and graphite as anode component [5]. This is also the scheme of the first commercial batteries, which entered the market at the beginning of the 1990s [5,6]; their diffusion made possible the development of new technologies, particularly of portable and wearable devices, and further drove the transition from traditional combustion engine-based transportation to electric transportation [1,7–12].

The rapid development of LIBs has led to increased production efficiency and lower costs for manufacturers, resulting in a growing demand for batteries and their application across various industries, particularly in different types of vehicles (Figure 2). The revolutionary impact of LIBs on our lives has been recognized with the Nobel Prize to Goodenough, Whittingham, and Yoshino in 2019 for the development of such technology. Then, the expansion of the market has resulted in a demand for the recycling of LIBs. In fact, the sales of electric vehicles reached the milestone of 1 million in 2017, and it is projected that more than 1 million electric vehicle batteries will reach their end-of-life by 2030 globally [9]. It is anticipated that around 14 million waste LIBs will be generated annually by 2040. Furthermore, in the near future, no revolutions in the LIB working scheme and materials are expected; thus, we will rely on the currently available technologies with demand growth of 14% Compound Annual Growth Rate (CAGR). Thus, the two problems of correct management of waste LIBs and the supply of critical raw materials needed to satisfy the always-growing demand for LIBs are becoming crucial.

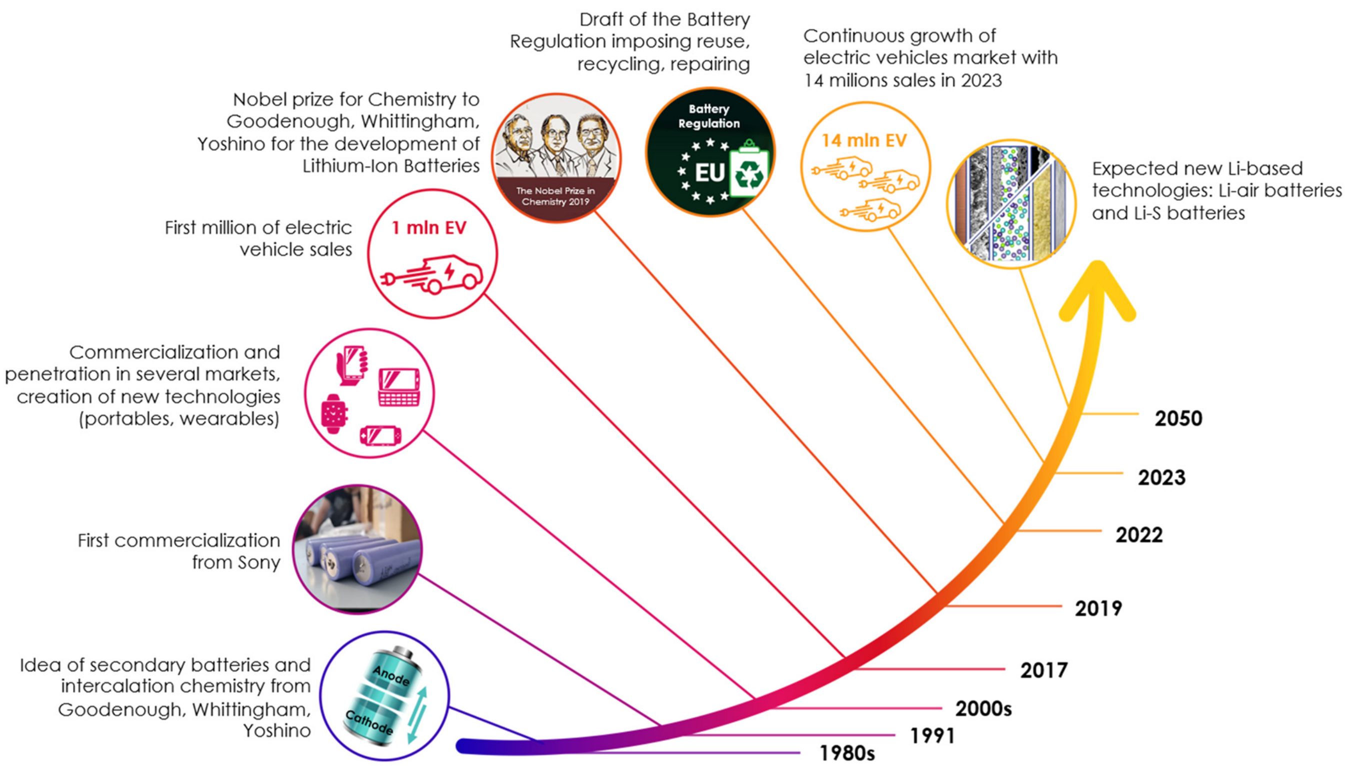


Figure 2. Milestones in the development of lithium-ion battery technology.

Indeed, LIBs are made of several materials, such as mixed crystalline oxides (containing lithium, nickel, and cobalt), organic solvents (flammable and explosive), polymers, and fluoride-based materials, and none of these should not be disseminated in the environment. Additionally, some of these substances are characterized by low natural availability (such as Li and Co) and/or can originate from states with low political stability (mainly Co). Table 1 shows the most diffused LIB cathode materials with their structures, main functional properties, and applications: $LiMn_2O_4$ (LMO), $LiCoO_2$ (LCO), $Li(Ni/Mn/Co)O_2$ (NMC) followed by numbers indicating the stoichiometric ratio among the Ni:Mn:Co, and $LiFePO_4$ (LFP).

Table 1. The most diffuse cathode materials, their structures, main functional properties, and applications. $LiCoO_2$ is usually referred to as LCO; $Li(Ni/Mn/Co)O_2$ is referred to as NMC followed by numbers indicating the stoichiometric ratio among the Ni:Mn:Co; when Mn is substituted by Al, it is referred to as NCA, $LiMn_2O_4$ as LMO, $LiFePO_4$ as LFP.

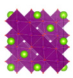
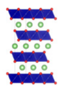
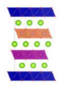
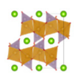
	$LiMn_2O_4$/LMO	**$LiCoO_2$/LCO**	**$Li(Ni/Mn/Co)O_2$/NMC**	**$LiFePO_4$/LFP**
Structure				
Specific capacity/mAhg-1	148	275	275	170
Practical specific capacity/mAhg-1	120	140	160	150
Average discharge potential vs. graphite/V	4.0	3.8	3.9	3.5
Energy density/WhKg-1	480	564	608	525
Application	Power tools, electric bikes	Portable devices	Portable devices, electric vehicles	Power tools, large electric vehicles
Market share	Small	Dumped	Dominant	Expanding

Table 1. *Cont.*

	$LiMn_2O_4$/LMO	$LiCoO_2$/LCO	Li(Ni/Mn/Co)O_2/NMC	$LiFePO_4$/LFP
Global evaluation	Medium safety, low cost, low lifetime, medium energy density	Low safety, high cost, medium performance	Medium safety, medium cost, high energy density	High safety, medium cost, medium energy density, high thermal stability

Climate-impacting emissions are generated during battery production, but they can be minimized by reusing the materials [13–17] at an industrial level from different companies patchily around the world such as Umicore, Ecobat, GEM, and HUAYOU Cobalt; however, the recycling of LIBs is often deemed inadequate, unlike lead-acid batteries that are usually recycled over 90% of the time in Europe. The main recycling facilities are based in China, South Korea, and Japan (not surprisingly, the main LIBs producers), while from the European point of view, the situation is particularly alarming as no systematic recycling paths and plans are present.

Current research on LIB recycling is extremely active and diversified, dealing with all the aspects of such a complex topic. A schematic representation of the LIB life path is reported in Figure 3. The creation of dedicated paths for collection from end users, facilities for the recovery of w-LIBs from the disposed devices, discharge, disassembly of the battery packs and/or battery units, and analysis of the state of health and state of charge of the waste LIBs are necessary [18–26]. Reuse for a second life and less demanding applications (such as static applications) can prolong the life of the battery. When reuse is not possible, the recovery of single components (cathode and anode materials mainly) has been proposed for their healing and use in the assembly of new batteries [18–21]. If healing is not possible due to the severe degradation of the considered components, recycling for the recovery of critical raw materials is the desirable solution; the recovered critical raw materials can be implemented into the production of new components for the assembly of new batteries, dumping the pressure on the extraction of these elements from natural sources [22–26].

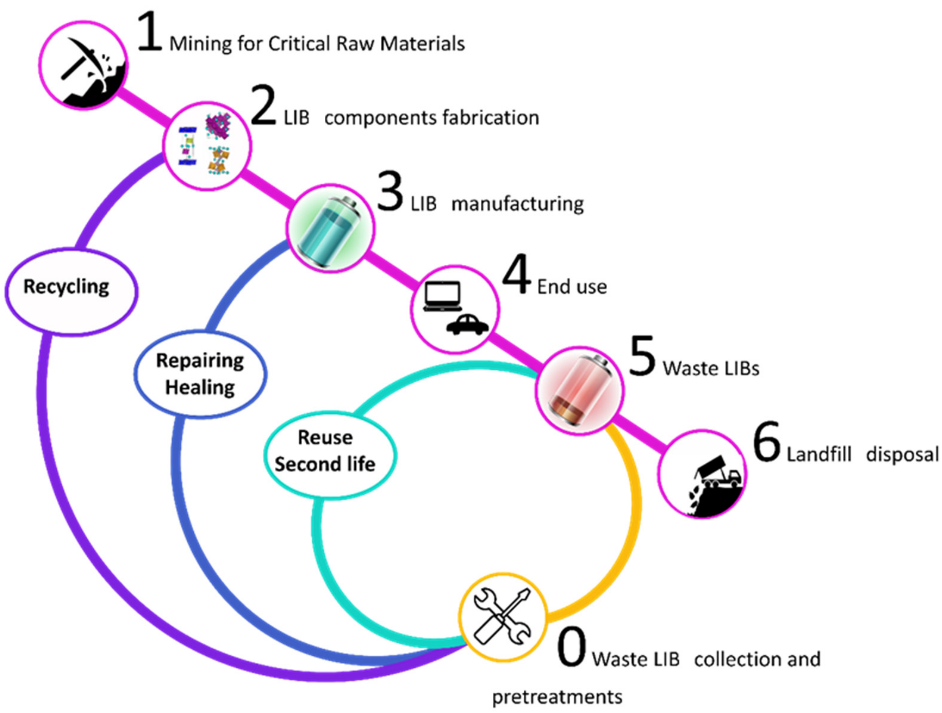


Figure 3. Scheme of the LIB lifespan, from production to disposal (steps 1–6, colored in pink) as implemented today, according to a linear economy scheme [7,8,13,16,27–40]. To switch to a circular economy scheme, some preliminary complex steps are needed (step 0, in yellow).

Due to the intrinsic complexity of the topic and the variety of approaches, we here report the most significant works and reviews addressing specific challenges and problems in the LIB value chain.

Today, scientific research mainly focuses on the methods of extracting, healing, and/or recycling cathode materials. Several types of recycling pretreatment processes are discussed in this review, reporting on their technological development and status [27–33]. There is a significant focus on undertaking a thorough assessment of the current hydrometallurgy technology for extracting valuable metals from waste LIBs, in comparison to the presently developed pyrometallurgical recovery schemes. For instance, several recent reviews are dealing with the specific aspects and steps reported in Figure 3, ranging from pretreatments, discharging and disassembly, recycling, reuse, and repairing [18,19,28,31,33–39].

Major efforts are focused on the recovery, recycling, and/or repairing of cathode materials due to the high concentration of valuable critical raw materials [19,39–42]. On the contrary, recycling anode materials and electrolytes has been an area that has not received as much attention as the recycling of other battery components [43–47]. This is a significant issue because the recovery of these materials is crucial to closing the materials loop and achieving a sustainable battery economy [48]; these aspects will be discussed in the next paragraphs.

Moreover, the potential environmental impact of the recycling process has not been given sufficient attention in most studies. There are concerns about the negative impact of the process on the environment due to the generation of toxic and hazardous waste materials during the recycling process. Thus, it is necessary to research the environmental implications of LIB recycling to ensure that the process is sustainable and environmentally friendly [49].

This document's primary objective is to offer a thorough review of the current knowledge concerning the recycling of materials from LIBs. The goal is to outline the current recycling chain and explore the factors that impact volumes, processes, and legislation. Significant attention is also given to sustainability analysis methodologies in recycling processes. Following this, a summary of existing research in this field is presented to encompass available insights and identify areas where knowledge is deficient. The aim is to pinpoint current knowledge gaps and tackle technical challenges requiring resolution to enhance the efficiency of the LIB recycling chain. This is very important also given the recently developed pyrometallurgical technologies that are able to increase the Li recycling ability, which was hard to recover in the past [50,51]. The aim is to address the work of the most recently developed technologies, which are still not industrially available [50,52].

Finally, the conclusions drawn from this study are summarized and contextualized concerning the Swedish Energy Agency's work towards achieving the United Nations Sustainable Development Goals (SDGs), specifically SDG 7 (Affordable and Clean Energy), SDG 12 (Responsible Consumption and Production), SDG 13 (Climate Action), SDG 14 (Life Below Water), and SDG 15 (Life on Land). This study also aims to contribute to the battery industry's efforts to implement these SDGs.

2. Recycling of Spent Li-Ion Batteries: Study Design

Figure 4 shows the scheme of the different approaches developed after the pretreatment of the LIBs for the different classes of components.

Spent lithium-ion batteries contain significant amounts of high-value elements, such as cobalt (5–20 wt%), lithium (5–7 wt%), and nickel (5–7 wt%), which are present in higher concentrations than in natural ores [53].

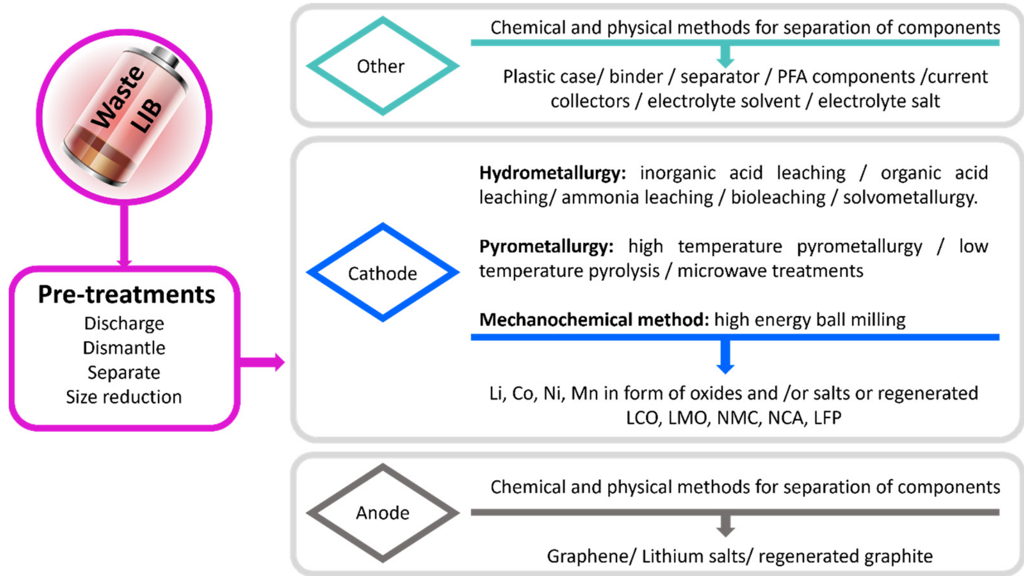


Figure 4. Scheme of the different approaches developed for the different classes of components.

To analyze the evolution of research interest in various recycling methods since 2018, a total of 170 review papers were assessed. According to the evaluation of these review papers, most publications (129) are related to the hydrometallurgical recycling route. The direct physical recycling route has 62 publications, while the pyrometallurgical recycling route has only 38 papers. The list of these papers with the corresponding keywords is available as Table S1 in Supplementary Materials. The significant level of research interest suggests that this recycling approach will likely become commercially viable soon. As reported in Figure 5, the NMC111 ($LiNi_{0.33}$ $Mn_{0.33}Co_{0.33}O_2$) battery is the most profitable type of electric vehicle (EV) battery to recycle. Indeed, battery recyclers could earn around USD 42 per kilowatt hour when recycling an NCM111 battery. In contrast, LFP batteries had the lowest salvage value at around USD 15 per kilowatt hour.

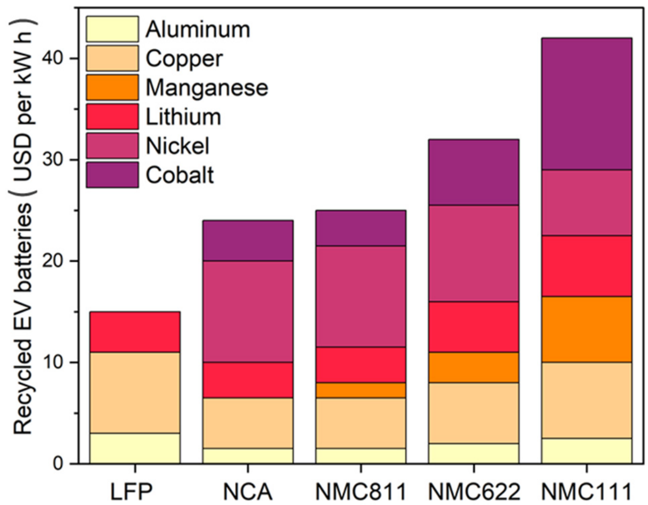


Figure 5. Value of recycled electric vehicle (EV) batteries in 2020 by battery type, by cathode chemistry (in USD per kilowatt hour) (In Statista. Retrieved 25 April 2023, from [54]). According to data from 2020, the most profitable type of EV battery to recycle was the NMC111 ($LiNi_{0.33}$ $Mn_{0.33}Co_{0.33}O_2$) battery. Battery recyclers could earn around USD 42 per kilowatt hour when recycling an NCM111 battery. In contrast, LFP batteries had the lowest salvage value at around USD 15 per kilowatt hour.

2.1. Materials Recycling

The minerals required in LIBs depend on the chemistry of the cathodes, but the key materials are lithium, cobalt, nickel, graphite, and manganese. In comparison to conventional vehicles, an EV requires six times more minerals, as shown in Figure 6. This means that the energy sector's demand for critical minerals could increase by up to six times by 2040. Recycling battery materials is therefore crucial for not only reducing waste but also for the future battery value chain.

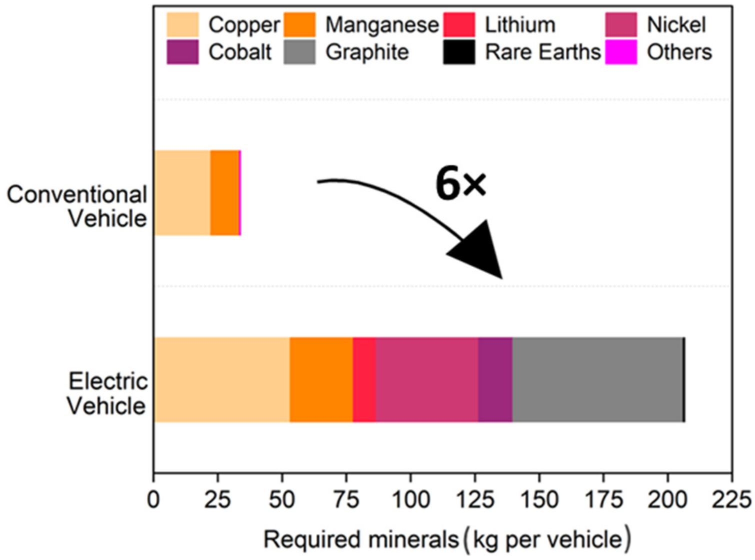


Figure 6. Amount of minerals present in battery electric vehicles compared to conventional vehicles, categorized by type, in kilograms per vehicle (data from 2022) [55].

Currently, almost all metals utilized in batteries, such as nickel, manganese, cobalt, and lithium, can be recuperated and recycled. However, more advancements are necessary to recycle other battery components such as graphite, liquid electrolytes, and plastic separators between the cathode and anode surface. This is an ongoing process that requires further development.

In the next sections, the main proposed recycling technologies are reassumed.

2.1.1. Pretreatments

The initial stage of the recycling process involves pretreatment, where the battery casing is separated from the valuable components [27–33]. Spent LIBs often retain a residual voltage, which, if left unaddressed, can pose a safety risk due to the potential for combustion or explosion. Discharge, battery disassembly, and sorting are typically involved in the pretreatment of waste LIBs. Following pretreatment, the waste batteries can be broken down into various components such as aluminum and copper foils, separators, plastic, and others. Manual disassembly involves carefully removing the battery shell with specialized tools while taking safety precautions to obtain the battery core coil, which can then be manually separated into its constituent parts, including the cathode, anode, and organic diaphragm. In contrast, mechanical processing can be performed on a larger scale and is more cost-effective, making it a more economically viable option. The most valuable component of a battery cell is black mass, and mechanical pre-treatment is primarily designed to achieve its optimal recovery and separation. To accomplish this, a sequence of mechanical processes, including crushing, sieving, magnetic separation, fine crushing, and classification, is typically performed in a specific order.

Widijatmoko proposed a flowchart for the recovery of black mass [56]. The cells underwent shredding and subsequent sieving to partition the components into various size fractions. The fine black mass was extracted from the coarse foils using an attrition scrubbing technique. Among the different size fractions, the 850 µm fraction was found to yield the most desirable black mass recovery composition with minimal copper and aluminum content during milling. However, a significant portion of the black mass remained bound together by the polyvinylidene fluoride (PVDF) binder [57].

2.1.2. Pyrometallurgy

Pyrometallurgy is a widely used recycling method that focuses on recovering valuable metal elements from spent LIBs, such as metals, metal oxides, or alloys [34,36]. The technology involves a metallurgical process that separates and extracts valuable metals from solid resources at high temperatures based on their melting and boiling points. The process is physical and chemical and comprises three stages: pyrolysis, metal reduction, and gas incineration. During the pyrometallurgical process, the organic components of LIBs are first thermally degraded, followed by the production of metal alloys using reducing agents at around 1500 °C. Finally, the gas is pyrolyzed and quenched at around 1000 °C to prevent the formation and release of toxic gas [58]. The pyrometallurgical process offers several advantages, such as a short process flow, low equipment requirements, and strong operability. It is also a mature technology and is currently the most diffused way to recycle LIBs in the available plants. However, the process also has some disadvantages. High energy consumption is one of the primary concerns associated with pyrometallurgy, as well as significant environmental pollution due to the release of pollutants during the process. The process also results in low product purity, and metals cannot be often recovered as single metals or single oxides, but as metal alloys [44].

One of the most significant challenges associated with pyrometallurgy is the difficulty in recovering lithium and aluminum during the reduction smelting process due to their vigorous reducing activity. The slag-forming agent also enters the slag phase, requiring further treatment. Overall, despite the challenges, pyrometallurgy remains a widely used recycling method due to its efficiency and effectiveness in recovering valuable metals from spent LIBs.

The carbothermic reduction can also be applied to recover metals from spent LIBs. Carbothermic reduction is a process in which a metal oxide is reduced to a metal using carbon as the reducing agent [59]. In the case of spent LIBs, the metal oxides that are present in the cathode material can be reduced to their respective metals using this process. The process involves mixing the spent battery material with carbon, typically in the form of coke, and heating the mixture in a furnace to high temperatures (usually around 1000–1200 °C). The carbon reacts with the oxygen in the metal oxide, forming carbon dioxide, and the metal is left behind. The metal can then be recovered and reused [60]. This step is generally followed by solvent extraction and chemical precipitation. In particular, combining pyrometallurgical technologies with hydrometallurgical processes is a common approach to recovering valuable metals from spent LIBs. Umicore, for instance, uses this method to first obtain Co-Ni-Cu-Fe alloy through the reduction and smelting of spent LIBs, and then utilizes hydrometallurgy to obtain high-purity single metals and compounds [61].

Several studies suggest that microwave irradiation can lower the temperature required for reactions, even in non-catalytic reaction systems, by several hundred degrees when compared to conventional heating systems [62].

Recently, microwave radiation has been proposed as a sustainable alternative to conventional pyrometallurgical processes for recovering Li, Co, Mn, and Ni from spent LIBs. This approach utilizes a hybrid heating mechanism obtained by combining microwave radiation with other heating methods [63]. The black mass obtained from spent LIBs contains graphite, which can cause heating due to the interfacial polarization of carbon atoms. Hybrid heating technology utilizes an external susceptor to achieve carbothermic reduction conditions in just a few minutes. A significant advantage of this method is that it does not require the separation of anode and cathode materials during the pretreatment process. It can also process a mixture of various discarded LIB materials, which is typical in industrial recycling plants that do not segregate spent battery types. Consequently, primary waste sorting before treatment is unnecessary, which is not feasible in hydrometallurgical processes intended for specific waste recovery.

2.1.3. Hydrometallurgy

Hydrometallurgy is a promising recycling method due to its low energy consumption and ability to recover a wide range of valuable metals. One of the main advantages of the hydrometallurgical process is the ability to recover metals individually, as opposed to the pyrometallurgical process, where metals are recovered as alloys. This allows for greater control over the purity of the final products, as well as the potential for greater economic value.

In the leaching process, various chemical reagents, such as acids or alkalis, are used to dissolve the metal ions from the battery waste. Acid leaching is a commonly used method for recovering valuable metals from spent LIBs, where sulfuric or hydrochloric acid is used to dissolve the metal ions [64]. Alkali leaching, on the other hand, uses a basic solution, typically sodium hydroxide or potassium hydroxide, to extract the metal ions [65–68]. Biological leaching involves the use of microorganisms to dissolve metals, while special solvent leaching utilizes specific solvents to selectively extract certain metals [69].

After leaching, separation and purification techniques are used to remove impurities and isolate the metal ions for further processing.

Solvent extraction is a process that separates metal ions from impurities in a two-phase system by using solubility differences between the metal ions and the solvent. This technique is used after metal leaching and helps to remove impurities like aluminum, copper, and iron to achieve the desired metal purity. Chemical precipitation is another method used for metal separation and impurity removal. The process involves adjusting the pH of the solution to precipitate different metals.

In the hydrometallurgy process, inorganic acids like nitric acid (HNO_3), phosphoric acid (H_3PO_4), hydrochloric acid (HCl), and sulfuric acid (H_2SO_4) are commonly used as leaching agents [70]. However, these acids can release toxic gases such as Cl_2, SO_3, and NO_x during the leaching process, and the waste acid solution requires neutralization with a strong base to prevent water pollution [71].

On the other hand, the use of organic acids (either alone or in combination) like malic acid ($C_4H_6O_5$) [72], oxalic acid ($C_2H_2O_4$) [73], citric acid ($C_6H_8O_7$) [74], and formic acid (CH_2O_2) is considered more environmentally friendly. Organic acids can act as chelating agents, precipitants, and even reducing agents during the leaching process [75].

The efficiency of leaching Li, Mn, Co, and Ni from various cathode materials of waste LIBs, including NMC, LCO, and LMO, is influenced by both the process temperature and time. As the temperature increases, the efficiencies also tend to increase due to the endothermic nature of cathode material dissolution. The effect of time on the leaching efficiency depends on factors such as the type of leaching agent, leaching temperature, and the type of cathode material. In general, longer leaching times tend to improve the leaching efficiency, up to a certain point where further increases in time do not significantly improve the efficiency [76].

One potential disadvantage of hydrometallurgy is the generation of large amounts of wastewater containing chemical reagents and impurities, which require treatment and disposal. Additionally, the recovery of certain metals, such as lithium, can be challenging due to their high solubility and reactivity. Overall, however, hydrometallurgy offers a promising approach to the sustainable recycling of spent LIBs.

2.1.4. Biometallurgy

Bioleaching is a bio-hydrometallurgy method that employs microorganisms such as fungi, chemolithotrophic bacteria, and acidophilic bacteria as leaching agents to extract valuable metals from a substrate. These microorganisms use ferrous ions and sulfur as energy sources to produce metabolites in the leaching medium that facilitate the recovery of metals [77].

Selective microbial bacteria can be employed to facilitate the leaching of valuable elements through specialized metabolic processes [78]. One example of a microorganism that can be used for bioleaching is *Aspergillus niger*, a haploid filamentous fungus. *A. niger* facilitates bioleaching by secreting low molecular weight metabolites, such as organic acids, that dissolve metals from batteries. To extract Co and Li from spent LIBs, the spent medium bioleaching method was employed using the organic acids produced by *A. niger* [79].

Compared to traditional metal recovery methods, bioleaching presents several benefits, including complete metal recovery, simplicity, cost-effectiveness, and lower energy consumption. One of the significant advantages of bioleaching is that it does not require harsh conditions or specialized industrial equipment, making it an attractive technology for metal recovery. However, the slow kinetics of the process are a potential limitation. Although bioleaching is a promising technique, it takes longer to recover metals than other methods, which can be a disadvantage in terms of processing time and efficiency. Nevertheless, ongoing research in this field is focused on improving the kinetics of bioleaching and making it more efficient for industrial applications [80]. Due to the requirement for microorganisms to adapt and undergo genetic modification to withstand the toxicity of the leachate media from spent LIBs, as well as the relatively slow kinetics of the process, bioleaching is not currently well-suited for large-scale applications [77].

2.1.5. Solvometallurgy

Solvometallurgy is an extremely recent approach developed for the recycling of w-LIBs as an alternative to the hydrometallurgical processes and to overcome their main limitations. Indeed, as discussed in the previous paragraphs, hydrometallurgy exploits water-based solutions of leaching agents with inorganic and organic acids as the most reported for industrial and laboratory scale research, respectively [6,81,82]. This leads to the creation of large volumes of wastewater as, typically, the solubility of the leached metals is low in this water-based solution. Moreover, in the case of inorganic acid leaching, the formation of hazardous gas species such as NO_x, SO_x, and HCl need to be accounted for as a source of secondary pollution [6,82,83]. Solvometallurgy is based on the use of alternative leaching systems such as ionic liquids and deep eutectic solvents (DESs); particularly, this last class of compounds has led to appealing results since the first report of their exploitation for cathode recycling [84–87].

Generally, DESs are based on biodegradable and inexpensive components; from a structural and functional point of view, they present medium-high viscosity, low volatility, non-flammability, extremally low toxicity, and high thermal stability, thus presenting a desirable combination of favorable characteristics as possible leaching systems for w-LIBs. Indeed, due to their peculiar composition, the solubility of metals in the DES is generally high and, thus, the production of wastewater is reduced or eliminated [85,88–92].

DESs formally can be classified as a class of ionic liquids, and they are made of two or more components forming a eutectic mixture at room temperature. The condition to obtain a eutectic mixture is to combine a hydrogen bond donor (HBD) and a hydrogen bond acceptor (HBA) [93]. Recently, the general formula has been proposed as Cat+ X- zY with Cat+ as the cation, X- as the Lewis base, and Y as a Broensted acid; concerning this, a classification of DESs to rationalize the variety of compositions has been proposed [81,93,94]. Almost all the DES compositions that appeared in the literature for applications in the recycling of cathodes from waste-LIBs contain choline chloride as HBA and belong to Type III according to the classification reported above [93]. Several reports have appeared in the literature exploring several HBDs, reporting high efficiency for the leaching of LCO, NMC, and LMO materials [84,92–100]. In minor extension, DES has also been used for the detachment of electrodes from the current collector and anode recycling [101,102]; today, most studies are focused on the degradation of cathode components.

The range of explored conditions is wide, with the main parameters affecting the leaching process being the time (60 min–48 h), temperature (80–240 °C), and the liquid–solid ratio [81,84,86,93–100]. The mechanism driving the leaching is still not completely clear; the presence of a reducing agent seems to be necessary to force the Co^{3+}/Co^{2+} reduction and favor the degradation of the LCO materials [93–95,103]. The reducing ability of the HBD has been assessed through density functional theory (DFT) analysis and Capacitance-voltage (CV) measurements, but this kind of study on the leaching mechanism is still in its infancy. Several studies report the possibility of recovering the DES systems after the leaching process and propose its reuse for several leaching cycles; this aspect must be further investigated and combined with the determination of the leaching mechanism. Indeed, if the leaching reaction is driven by a reductive decomposition of the cathode material, the oxidation of the DES must be inferred. Thus, for a full recovery of the DES and its reuse, this oxidation must be reversed or compensated with a change in the DES composition. As a perspective, beyond the promising results obtained up to now in the high-yield degradation of cathode components, some efforts are needed to understand and rationalize the effect of the DES composition on the leaching process and to obtain insight into the leaching mechanism. This can lead to a further optimization of compositions and conditions for a more effective leaching process. As already stated, only small efforts have been made in the field of exploitation of DES for the detachment of electrodes from the current collectors and for the recycling of the anodes; also, these topics can be of high relevance in the future.

Overall, the different developed technologies here described present both advantages and weak points, as schematized in Table 2. Indeed, pyrometallurgy is the mainstream method at the industrial scale, and is the most diffuse and mature, with high recycling volume capacities and extremely simple operations. At the same time, it suffers from some severe limitations, the main one being the possibility to recover only a fraction of elements of interest. Indeed, with the technologies implemented today, it is not possible to recover lithium, considered one of the most critical and strategic elements, while also considering the new regulation proposal from the EU that makes Li recycling and reuse mandatory [104]. Hydrometallurgy can encompass this limitation if not associated with preliminary pyrometallurgical steps (as often implemented) but as an alternative independent strategy. Although still limited, it is already industrially exploited and is expected to grow rapidly thanks to this particularly appealing factor. The main drawback of such an approach is the creation of large amounts of wastewater due to the limited solubility of the relevant metals in water-based solutions [105]. Solvometallurgy represents, in this sense, a possible and appealing alternative as it allows for low-temperature operation (80–250° typically), complete dissolution of the cathode materials, and high yields of recovery of different elemental species.

Table 2. Direct comparison of the main recycling technologies described in this review.

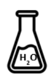
					
	Pyrometallurgy	**Hydrometallurgy**	**Biometallurgy**	**Solvometallurgy (Ionic Liquid)**	**Solvometallurgy (DES)**
Advantages	Short process flow, low equipment requirements, strong operability	Low energy consumption, great versatility, high product purity, high recovery efficiency	Complete metal recovery, simplicity, cost-effectiveness, low energy consumption, mild conditions	Nonflammable, low volatility, tunable	Nonflammable, low recovery cost, green process, cheap and easy preparation, low toxicity
Disadvantages	High energy consumption, poor metal purity, difficulty in lithium recovery	Need to dispose of large amount of acid and toxic wastewater, long recovery process	Long processes and low kinetics, vulnerability to pollution	Expensive	Difficulty to scale-up, low cathode/DES ratio

Table 2. *Cont.*

	Pyrometallurgy	Hydrometallurgy	Biometallurgy	Solvometallurgy (Ionic Liquid)	Solvometallurgy (DES)
Applied at industrial level	Yes	Yes	No	No	No
Main source of pollution	Emission of polluting gases and production of slags	Release of toxic gases (e.g. NO_x, SO_x, Cl_2)	-	-	-

2.1.6. Direct Lithium Supplementation

Direct lithium supplementation is a method of recycling the cathode of LIBs that involves replenishing the lithium content in the cathode material to restore its capacity and cycling performance [106]. This method aims to address the issue of lithium deficiency in spent LIBs, which can lead to a decrease in the overall performance of the battery. Direct cathode regeneration methods have been proposed as a means of closed-loop recycling, which can mitigate raw material shortages and supply chain risks [107]. The regeneration process involved supplementing metal ions, granulation, ion doping, and heat treatment, generally resulting in excellent electrochemical performance. One study proposed the use of an eutectic LiI-LiOH salt with a low eutectic point to create a Li-rich molten environment, which offers excess lithium and benefits ion diffusion compared to a solid environment [106]. This eutectic salt, combined with additives, simplifies the recycling process and endows the cathode materials with lithium supplementation and structural ordering, leading to the restoration of capacity and stable cycling performance. Another study developed a green, efficient, closed-loop direct regeneration technology for reconstructing the $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ cathode material from spent LIBs [108].

Nevertheless, the applicability of the approach is constrained when faced with diverse cathode chemistries and degradation scenarios observed in different spent LIBs. Nearly all documented direct recycling techniques prove effective solely for one or two categories of spent cathodes characterized by a limited degree of degradation [106]. Specifically, these are instances where the cathode materials exhibit a high residual capacity and minimal structural damage. Existing direct recycling methods, however, largely neglect highly degraded cathodes marked by low residual capacity and significant structural defects, primarily due to the challenges associated with their repair.

2.1.7. Anode and Electrolyte Recovery

In recent years, there has been increasing attention given to anode recycling, although it has not yet been established to the same extent as cathode recycling. Anode recycling is becoming an increasingly important aspect of LIB recycling. At present, anodes for LIBs can be manufactured using a variety of materials, such as natural or artificial graphite, carbonaceous materials, or even silicon. Among these options, graphite is the most widely used material in commercial applications due to its excellent conductivity, stability, and low cost. In fact, graphite is the most applied material in commercial anode production processes [109]. Several approaches have been reported in the literature, including pre-treatment, pyrolysis, hydrometallurgy, supercritical, and water treatment. Recycling anodes involves removing the active material (e.g., graphite) from the copper or aluminum foil substrate. One common method is to use pyrometallurgical processes to treat the anode material at high temperatures, which oxidizes the carbonaceous material and leaves behind a mixture of metal oxides. This mixture can be further processed using hydrometallurgical techniques to extract valuable metals such as lithium and cobalt.

Another method involves the use of mechanical processes, such as grinding and sieving to separate the anode material from the substrate, followed by further processing using hydrometallurgical techniques [110].

Anode recycling has the potential to reduce the demand for virgin graphite and other raw materials, as well as decrease the environmental impact of producing new anodes. However, the challenge with anode recycling is that the graphite particles can become contaminated with metals and other impurities during the cycling process, which can affect the performance of the recycled anode material.

Like anodes, the recycling of electrolytes has gained attention in recent years. Typically, electrolytes are evaporated or burned during thermal processes, but various methods have been proposed to recover lithium from this component [111]. The initial technique involved in the recovery of the electrolyte was liquid extraction. Subsequently, a method utilizing sub- and supercritical media was proposed, which exhibited relatively high recovery rates. This method was also utilized for the retrieval of binders [112]. However, alternative methods for valorization have also been reported [113].

2.1.8. Current Collector Recycling

Current collectors, such as Al and Cu foils, are irreplaceable components of LIBs and have a significant impact on their performance. The recycling and reusing of these current collectors can contribute to reducing total global emissions and the demand for new materials [114]. Several strategies have been proposed for the separation and recovery of current collectors from spent LIBs. Pyrolysis and physical separation have been studied as effective methods for the recovery of valuable materials, including current collectors [115]. Another study developed a physical separation process using thermal and mechanical treatments to recover active cathode materials from current collectors [116]. The ultrasound-assisted Fenton reaction has also been explored for the selective removal of binders to recover cathode materials from current collectors [117]. Additionally, a solvent-based recovery process has been developed for the low-temperature and efficient separation of electrode materials from current collectors without damaging the active materials or corroding the metal foils [118]. These studies demonstrate various approaches to recycling and reusing current collectors in LIBs, contributing to the sustainable development of LIBs and the electric vehicle industry.

3. Sustainability

Recovered materials from spent LIBs can be reused in various fields, such as the production of new batteries, electronic components, and other energy storage devices or applications [119]. In some cases, recycling processes may have a lower environmental impact compared to extracting metals from the respective ores [120,121].

Table 3 reports some possible outputs of LIB recycling processes. The recovered metals, such as cobalt, nickel, and lithium, can be reprocessed and used in the production of new batteries, while other materials like graphite can be utilized in the production of lubricants and carbon-based products. Additionally, recovered materials can be utilized in the manufacturing of ceramics, glass, and other industrial materials. The reuse of these materials not only conserves natural resources but also reduces the environmental impact associated with the extraction and processing of virgin materials. Despite the possibility of proposing various alternatives to substitute raw materials with secondary materials, a significant disparity exists between research efforts and the practical potential for reusing these secondary materials. This incongruence persists even though these materials are classified as "sustainable," denoting their typical origin as recycled from waste [122].

Table 3. Potential outputs from recycling procedures [123].

Output Material	Definition	Comment
Active material	Lithium-ion batteries consist of both cathode and anode materials, with graphite being the predominant material used for the anode. The active material in the battery is a combination of these two components.	The output material represents a complete process for physical recycling directly.
Cathode material	Common cathode materials are $LiCoO_2$ (LCO), $Li_aNi_xCo_yMn_zO_2$ (NMC), $LiMn_2O_4$ (LMO), $LiNi_xCo_yAl_zO_2$ (NCA), and $LiFePO_4$ (LFP).	The direct physical route yields a purified output material.
Alloy and Slag	Typically, a pyrometallurgical process results in the production of metal alloys as the main product, with slag being a secondary byproduct of the process.	A final product that represents the entire process of pyrometallurgical recycling.
Salt of transition metals	Precipitation product in a hydrometallurgical process.	A final product obtained from a complete hydrometallurgical recycling process.
LIBs	Lithium-ion batteries are rechargeable batteries that comprise an anode and cathode, with an ion-conducting electrolyte present between them for the migration of lithium ions.	The highest level of refinement achievable in a recycling process is the production of a new LIB.

Then, the sustainability of the proposed procedures must be evaluated in detail to verify the convenience of the proposed recycling procedures.

3.1. Environmental Aspects

The environmental impact associated with LIBs, especially in electric cars, is a topic frequently covered by the media. Comparisons are often made with traditional fuel-powered vehicles or alternative technologies like fuel cells. In this frame, the end-of-life stage of the batteries is a crucial factor in determining their impact, as efficient recycling and reuse in second-life applications can potentially improve their overall impact, including that of electric cars. Conversely, inefficient recycling would have the opposite effect.

Overall, monitoring and managing emissions during the recycling process is critical to ensure the environmental sustainability of LIB recycling. It is essential to collect quantitative data on the emissions and to explore treatment options to minimize their impact on the environment and human health. Then, the environmental impact of the recycling processes is an important factor to consider, especially regarding air pollution, wastewater, and waste [124].

Emissions from technical processes can occur in two ways when recycling LIBs: reaction emissions and material losses. Reaction emissions are generated from chemical reactions during the process, such as thermochemical reactions [125]. Material losses, on the other hand, result from the release of components of the batteries during the recycling process. Both types of emissions can have environmental and health impacts, making it essential to monitor and manage them during the recycling process.

However, information on the complete process chain for LIB recycling is often limited, and only data on individual processes are available [126]. It is crucial to have quantitative estimates of the emissions to better understand the potential impact on the environment and human health. For air pollution and wastewater, it is necessary to know the pollutant, its concentration, and the volume flow of the polluted fluid per ton of recycled LIB. For solid waste, the specific type of waste and its mass per ton of treated LIB are essential to determine the potential environmental impact.

It is also important to consider treatment options for the resulting pollutants. For example, air pollutants can be treated using air pollution control devices, while wastewater can be treated using biological or chemical processes. Solid waste can be treated by various methods such as landfilling, incineration, or recycling. Finding the best treatment option will depend on the type and concentration of the pollutants, as well as the local environmental regulations.

The recovery and purity of valuable materials are closely related to the prevention of emissions, as material losses can contribute to pollution.

A mass balance analysis reveals that air pollution, wastewater, and solid waste are significant issues associated with pyrometallurgical recycling, while the hydrometallurgical route generates high volumes of wastewater [50]. However, the available literature provides mainly qualitative information on this topic. One reason for this is that emission monitoring is often not performed in lab-scale processes, as the upscaling can alter the amount and concentration of emissions. Nonetheless, these emissions have a significant impact on climate targets and can be the decisive factor in selecting the appropriate recycling route.

3.2. Economic Aspects

A circular economy seeks to manage waste by promoting reuse, responsible manufacturing, and recycling, while also reducing emissions and increasing natural resource efficiency. Achieving recycling goals and proper battery disposal requires an understanding of the value chain and recycling routes of LIBs. Additionally, recycling processes must be economically feasible. Currently, China dominates the recycling industry with a 73% global recycling capacity share, despite having only 45% of domestic LIBs available for recycling. However, as the volume of discarded LIBs increases with the rise of EVs and other applications, recycling will become more financially attractive worldwide. Tax exemptions or reductions can make recycling economically viable, promote circular economy practices, and drive job creation and economic development in the areas where recycling plants are built [127].

Collaboration through joint ventures is a critical aspect that must be taken into account in achieving recycling goals. Joint ventures can incentivize companies to work together towards a common goal, ultimately enabling the achievement of circular economy objectives. By facilitating the entire value chain, including logistics, transportation, and battery disposal, joint ventures can help overcome barriers to battery recycling.

StartUs Insights Discovery Platform [128] has identified five prominent battery recycling startups with the potential to significantly impact the market and generate substantial revenue. Among these, Li-Cycle is a Toronto-based recycling company that specializes in recovering minerals from LIBs while minimizing greenhouse gas emissions. The company's capacity is expected to increase tenfold in just four years across its North American and European facilities (see Figure 7) [129].

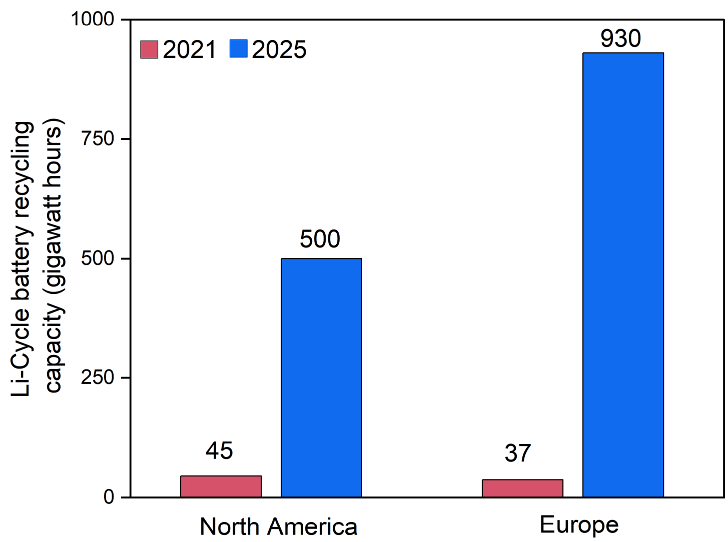


Figure 7. Li-Cycle's lithium-ion battery recycling capacity in gigawatt hours (GWh) for 2021 and projected capacity for 2025 by region (in gigawatt hours).

3.3. Technologies Sustainability Evaluation

Technology Readiness Levels (TRL) are utilized to evaluate the level of maturity of a given technology. Projects are assessed against specific criteria for each of the nine TRL levels, with a TRL rating assigned based on the project's progress. TRL 1 is the lowest level, and TRL 9 is the highest.

Life Cycle Assessment (LCA) may be used to find the environmental benefits that battery recycling may obtain [130].

Various life cycle analyses have been carried out on batteries, utilizing different comparisons and approaches. The initial studies, published in the early 2010s, compared LIBs with other battery chemistries, such as nickel cadmium, nickel metal hydride, and lead-acid batteries [131]. However, many of these studies rely on limited data and are often supplemented with calculations based on lab processes.

There are significant discrepancies in the values obtained from various life cycle analyses of batteries, which can be attributed to various factors, such as differences in energy mix values and types of data used. One reason for the differences is that different studies have different functional units and purposes, such as comparing different battery chemistries. Additionally, the electricity source considered can also impact the results, as the use of US or Chinese electricity for processing 1 kg of waste batteries can significantly increase greenhouse gas emissions and energy consumption. Furthermore, most studies have only covered cradle-to-gate or cradle-to-wheels stages, with other life cycle steps not being discussed.

The unclear benefits of pyro- and hydrometallurgical recycling process steps are why recycling LIBs does not always seem advantageous [132].

Life-cycle assessments (LCA) that measure cumulative energy demand and global warming potential suggest that the recycling of the most abundant materials in LIBs, such as Al and Cu, offers the most significant environmental advantages. Recovering critical materials such as Li, Co, and Ni shows fewer benefits in comparison. However, there are four significant limitations to LCA analysis for waste recovery. These include unspecified system boundaries, missing information, unconvertible data, diverging development states, and low affordability of studies conducted on low TRL technologies [61].

A very recently published paper recommends some strategies for conducting LCA of batteries to improve their interpretability, representativeness, and impact, and to increase their significance [133]. For example, it is suggested that LCA focus on long-term trends in resource depletion rather than treating known reserves as a fixed quantity and that different battery manufacturing facility scales are explored to capture size- and throughput-dependent impacts.

Further research is necessary for a comprehensive assessment of recycling routes, including the development of databases and methodologies to also allow for the evaluation of recycling processes developed only on a laboratory scale. The LCA methodology must also incorporate waste management and critical material recovery perspectives. To achieve this, different functional units and harmonious system boundaries should be evaluated, and a life-cycle assessment developed. The challenge of different products resulting from recycling routes must also be addressed. These aspects are complex, but a sustainability evaluation is crucial to assist decision-makers in making accurate decisions.

As a result, some examples of simplified tools for sustainability analysis have been developed [134]. A novel approach called ESCAPE (Evaluation of Sustainability of material substitution using CArbon footPrint by a simplifiEd approach) has been introduced to aid in the effective and sustainable utilization of raw materials, with the ultimate goal of preserving primary raw materials [135,136]. The ESCAPE approach offers a significant benefit in that it enables the assessment of technology sustainability, even if the technology is only in the laboratory phase. This validation process helps accelerate the technological transfer of recycling technologies, which is particularly relevant for LIBs since recycling technologies are still in their infancy. Through a screening approach, the ESCAPE method was recently employed to propose and examine the sustainability of 33 distinct technologies available in the literature for LIB recovery, despite being only in the laboratory phase, as a preliminary step before conducting a full LCA analysis [123,137]. The studies illustrate the diverse elements that affect the sustainability of technologies, including the utilization of thermal and mechanical treatments, chemicals, and water. The results, which are in line with the available data from life cycle assessments, indicate that chemicals and ultrapure water consumption are the most energy-demanding processes. This emphasizes that pyrometallurgy could be a more ecologically responsible choice when compared to hydrometallurgy, as it has the potential to lower both the carbon footprint and energy usage.

4. Open Issues

The recycling of LIBs is not without its challenges and implications, and these need to be considered at various levels of policy, industry, and research. One of the critical issues is the growing number of different types of battery chemistries, which makes it challenging for recyclers to accurately sort and classify batteries. This is because each chemistry type requires a specific recycling process, and improper sorting could lead to inefficient recycling, safety hazards, or even harm to the environment [138–141].

In addition to sorting, the disassembly of battery modules and cells is a complex and time-consuming process [142–145], particularly for hydrometallurgical recycling methods [70,146]. Moreover, the flammability of LIBs poses a safety concern during the disassembly process, as they can catch fire or explode upon exposure to mechanical stress [147].

Unfortunately, research on sorting, disassembly, and discharge of batteries is relatively rare, despite these safety concerns. Therefore, it is crucial to address these issues to ensure efficient and safe recycling of LIBs [148–150].

Another area where research is lacking is in the design of batteries for reuse and recycling. Currently, most batteries are designed without considering their end-of-life management, which makes recycling difficult and costly. Therefore, it is necessary to develop battery designs that take into account their recyclability and ease of disassembly [151].

Moreover, there is a need to focus on the pre-treatment and discharge of batteries, as these steps can significantly impact the efficiency of the recycling process. Currently, there is a lack of research in these areas, and more attention needs to be paid to improve the quality of the recycled outputs.

Finally, safety, work environment, and transport are essential considerations in the overall research on LIB recycling [152–155]. These aspects have not been given much attention, despite their significant impact on the industry's sustainability.

To achieve high recycling rates at a competitive cost and to meet the growing demand for LIBs, significant innovation is needed in integrating existing recycling methods for LIBs [156,157]. This will require a collaborative effort from policymakers, researchers, and industry players to overcome the challenges and make recycling more efficient, safe, and sustainable.

4.1. Batteries Collection

The collection and proper recycling of electronic waste is currently insufficient, with only a small fraction being properly managed. According to the Global E-Waste Monitor 2020 report, Europe has the highest rate of collected and recycled e-waste at 42.5%, followed by Asia at 11.4%, the Americas at 9.4%, Oceania at 8.8%, and Africa at 0.9%. In total, only 17.4% of the electronic waste generated worldwide is documented to be collected and recycled [158].

A successful recycling strategy for end-of-life batteries requires an efficient collection system. The number of spent batteries entering the recycling stream is determined by the collection rate, which is a key factor in the economic and ecological output of the recycling system as a whole.

However, the lack of collection is often considered a limiting factor for recycling, as individuals and companies tend to hoard used batteries for too long or dispose of them improperly in ordinary household waste.

Creating an effective collection infrastructure is a significant obstacle due to the diversity of battery types available in the market. Lithium-ion batteries are utilized in various applications, which leads to a vast range of battery designs differing in capacity, size, shape, and chemical composition [159].

Various collection systems need to be established due to significant differences in the markets. For example, the collection of smaller household batteries from electronic devices can be facilitated by placing containers at manufacturer and retail partner sites. In contrast, larger modules from EV and stationary battery energy storage devices necessitate disassembly and should only be handled by dedicated personnel [160].

Despite the potential benefits of direct recycling processes for LIBs, the current challenge lies in obtaining sufficient quantities of waste (black mass) for a single source, such as a laboratory. To overcome this issue, research centers, universities, and industries need to collaborate. Companies can facilitate a stable supply of spent LIBs while identifying technical obstacles in industrial production. To expand the scope of experiments and address technical concerns, universities and laboratories can conduct research. On the other hand, the research centers should experiment with new technologies at a larger scale than the laboratory ones and address technical issues.

By conducting both lab-scale and industrial-scale experiments on spent LIBs, the collaboration between industry, universities, and laboratories can break through technical barriers and ultimately support the realization of LIBs recycling facilities in increased TRL in comparison to the laboratory scale ones. This collaborative effort could significantly minimize pollution and maximize the benefits of recycling.

Future research should prioritize tackling the existing challenges that hinder efficient recycling, while also fostering a comprehensive understanding of the recycling system among all stakeholders.

4.2. Battery Disassembly

Recycling methods face a significant challenge due to the highly repetitive and labor-intensive nature of disassembly, particularly in mechanical procedures [37,38]. This is compounded by the fact that LIB cells come in cylindrical, prismatic, or pouch shapes, and variations in composition and shape due to the assembly into modules and packs create significant obstacles to recycling. Moreover, each manufacturer has a unique design or recipe, adding to the complexity of the process.

Automated disassembly is necessary to address the inefficiency and labor intensity of manual disassembly in recycling LIBs [161]. Artificial intelligence, machine learning, and deep learning can be utilized to develop automated or semi-automated disassembly methods based on the standard size and shape of battery packs, which can reduce costs and labor. To achieve automation, a robot can be employed for safe and efficient battery retrieval. Automatic dismantling processes, residual energy detection, secondary utilization, and chemical recovery are also crucial for the development of LIB recycling. However, the large variety of EV battery designs currently on the market poses a challenge for third-party reuse centers as they cannot plan ahead to optimize disassembly processes and determine the best second-life application [145].

4.3. Lack of Policies and Regulations

The successful implementation of government regulations and legislative management is crucial for the efficient and sustainable recycling of LIBs. It is essential to have globally implemented regulations that ensure the safe and effective handling of spent batteries. In China, the government has already implemented recycling policies for end-of-life EVs and waste LIBs. However, there is a need to enhance the collection rate of end-of-life spent LIBs to avoid potential pollution problems [162]. Furthermore, a recent study focusing on the policies of power battery recycling in China highlighted the concentration of recycling policies at the midstream processing end, while the upstream and downstream policies are comparatively deficient. The study also emphasized the lack of top-level legal construction and the imperfect recycling network, suggesting the need for modifications and relevant policy suggestions [163].

European governments and companies in the battery sector are making efforts to improve legislation on batteries and accumulators [164]. The Chinese experience provides valuable insights into the need for a holistic approach to recycling policies, covering all stages of the battery life cycle, from production to end-of-life management. Such policies should aim to improve collection rates, establish legal frameworks, and create incentives for echelon exploitation enterprises. Additionally, the focus on the entire life cycle of power batteries highlights the need for a systemic and integrated approach to recycling policies, ensuring environmental sustainability and resource conservation.

One important aspect of the regulations is the standardization of pack and module design, which can facilitate automatic recognition and disassembly. This can be achieved through clear external labeling that includes important information such as the manufacturer, manufacturing date, and battery type. Proper safety measures and regulations should also be in place to mitigate the risks associated with battery disassembly, such as flammable electrolytes and potential short circuits that could cause explosions or fires.

The development of effective regulations requires consideration of the technical differences in battery design and usage, as well as the immaturity of technology in this field. Developing guidance documents and expanding Extended Producer Responsibility systems while allowing for flexible implementation is suggested to facilitate the creation of a large-scale, efficient, and traceable LIB recycling management system in Europe [164]. The successful implementation of recycling regulations for lead-acid batteries could serve as a useful template for LIB recycling regulations. Increasing public awareness and education, establishing relevant organizations, and improving overall environmental literacy are also important steps towards successful implementation.

National-level recycling centers established by local governments, known as governmental recycling centers, can promote proper regulations and management of the LIB recycling market. They can improve recycling networks and increase recycling through regulated channels following relevant national laws. Although most countries do not have government-affiliated LIB recycling centers currently, they are expected to be established in the future.

To prevent spent LIBs from entering the black market and to create an organized recycling market, it is necessary to establish a battery-tracking mechanism. Each battery can be assigned an identification number, which can be uploaded into the tracking system throughout the end-of-life value chain to facilitate recycling development. This can also help to ensure that recycling processes comply with established regulations and standards. By implementing these measures, we can establish a sustainable and efficient circular economy for LIBs, which benefits both the environment and society.

4.4. Scaling-Up and Industrialization

Although there has been a recent surge in research on LIB recycling, many of the proposed methods are still limited to lab-scale experiments. In such experiments, the researchers have a greater degree of freedom in choosing parameters, chemicals, or processes, which may not be possible or are more challenging to implement at a larger scale.

Despite this, there are some examples of lithium-ion battery recycling being industrialized. Some of the main companies using the reported recycling methods (sometimes in combination with each other) include Umicore, Sony Sumitomo, Sumitomo-Sony, Retriev Technologies, Li-Cycle, Accurec, Recupyl Valibat, and Akkuser. These companies have varying recycling efficiencies, purity of recovered materials, and declared plant capacities [165].

In terms of development status, pyrometallurgical treatment is the most advanced. The current plants implementing this method have reached a technology readiness level of 9, indicating full commercial application and providing the highest recycling capacity available today [132].

The pyrometallurgical route has become the most mature and widely used method for LIB recycling because many companies are already involved in related fields such as mining and waste treatment. Therefore, they could leverage their existing company structures and equipment, such as furnaces, to adapt their established processes for the recycling of LIBs [132].

On the other hand, companies utilizing the hydrometallurgical route (with a technology readiness level of 9) are newly established solely to treat lithium-ion batteries.

Additional funding for pilot projects can help fill the gaps in data within the industry's recycling process, generate realistic data for investment planning, and obtain verified data that enable battery manufacturers to confirm the quality and compatibility of recycled materials. It is worth noting that some hydrometallurgical companies operate in dislocated hubs where they collect LIBs and mechanically separate the black mass. They then treat the black mass in a single hydrometallurgical facility, reducing transportation costs and the risk of fire [166].

In contrast, pyrometallurgy processes have the advantage of being able to handle small-format LIBs commonly found in portable devices, as different LIB compositions can be treated simultaneously, and batteries of different typologies can be easily managed. However, hydrometallurgy treatment requires prior mechanical treatments to select the black mass, which is usually performed in separate facilities within the same company or even by different companies.

5. Design for Recycling

End-of-life considerations have traditionally not been a significant concern in battery design, as the primary focus has been on optimizing performance. However, the recovery of critical raw materials (CRMs) in hydrometallurgical and direct recycling processes can be challenging, primarily due to difficulties in mechanical separations such as the sieving of crushed fragments [27]. The efficiency of sieving relies on the size distribution of battery fragments, which, in turn, depends on the efficiency of crushing. However, the greatest obstacle is the strong adhesion of active materials to current collector foils, which is caused by polymeric organic binders.

To promote the establishment of an effective economy of spent LIB recycling, an important notion is the concept of design for recycling. This approach involves also taking into account the possibility of recycling of battery cells when the batteries are designed. The design principles are primarily centered on three pillars: pack and module design, cell design, and material design [6]. Many larger batteries are composed of numerous individual cells organized in modules that are subsequently assembled into a pack. Enhanced separation of the battery components could result in more efficient recycling compared to conventional methods that involve shredding old batteries in their entirety.

However, manually disassembling such batteries is expensive, time-consuming, and poses significant safety hazards to workers [6]. Currently, achieving fully automated disassembly through self-learning robots is not feasible due to the intricate nature of certain disassembly procedures. However, a more viable option is the implementation of hybrid systems, where human workers collaborate with automated robotic arms to carry out the disassembly process.

Another example pertains to the objective of substituting polymeric organic binders with water-soluble binders, which can potentially be effortlessly eliminated during the recycling process by washing with water. Water-soluble binders may not provide the same long-lasting adhesion of the batteries' active substances to metal foils as the current polymeric organic binders.

The rational design of nanostructured hybrid materials based on carbon and metal oxides has shown promise in improving the kinetics of charge transfer and alleviating structural strain during charge/discharge processes [167]. Additionally, the improvement of the metal oxide-based electrode materials can be obtained by the use of materials with better cycling performance, such as nanosheets, nanorods, and nanospheres [168,169].

Designing for recycling also strives to standardize screw connections and junctions between modules or cells to simplify the automated disassembly of cells. A thorough understanding of cell chemistry is crucial since certain chemistries cannot be mixed during the recycling process [170].

Efforts in providing regulations devoted to battery classification, labeling, and coding are aimed towards achieving this goal, as the standardized reporting of details like LIB composition on labels affixed to the exterior of the batteries may aid in or facilitate the automatic sorting process [171].

In addition, the approach encompasses material design. Figure 8 summarizes the strategies for designing for recovery.

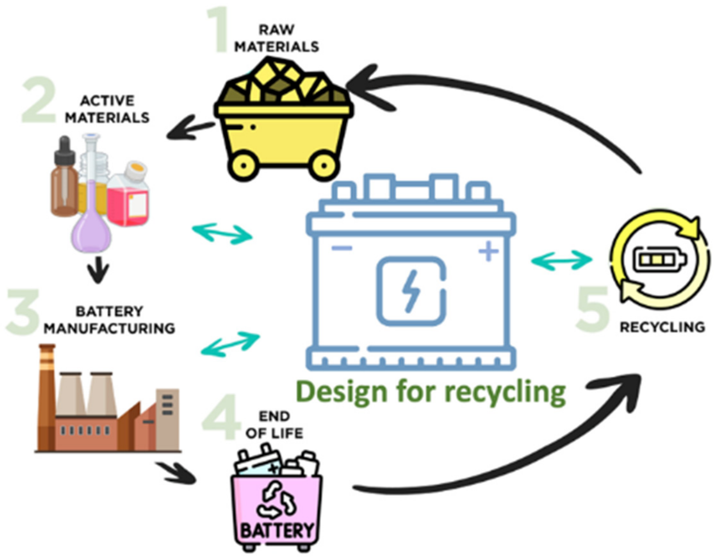


Figure 8. Design for recycling batteries must allow for (1) the use of recyclable materials, (2) information and easy access to data (for example cell chemistry), (3) ensuring and facilitating disassembly, and (4) facilitating materials recovery.

6. Future Perspectives

The prospects for LIB recovery have a focus on sustainable and efficient recycling technologies [172]. Direct regeneration and preparation of advanced materials are considered interesting strategies for cathodic upcycling, although challenges remain in industrial practices. Various approaches, including hydrometallurgical, pyrometallurgical, solvometallurgy, and biometallurgical methods, are being explored for metal extraction from spent LIBs, with different advantages and disadvantages related to high temperature and chemicals necessary for the reactions [51]. A challenge involves, for example, the substantial consumption of reagents/energy required to counterbalance the production of flammable and toxic compounds originating from fluorine-based substances, used as a binder [51]. They aim to restore the original components or convert them into low-carbon secondary materials, even if the sustainability of several of these processes is still quite low.

Moreover, the use of biomass pyrolysis gas-induced reduction and bioleaching approaches are being investigated as environmentally friendly and energy-saving methods for metal recovery. However, challenges such as controlling impurities, ensuring sustainability, and managing secondary wastewater need to be addressed. Hence, the enhanced recycling efficiency is more likely due to the characteristics of the input feed rather than the recycling process itself. This highlights that the potential of recycling processes is compromised by impurities and material losses during mechanical separations when dealing with mixed spent batteries [50]. Recycling companies are endeavoring to address challenges associated with LIBs pretreatments through methods such as additional crushing, thermal incineration, or dissolution of the binder in suitable organic solvents. However, none of these alternatives has proven to be optimal at this stage [50].

Overall, further research and development are needed to optimize recycling processes, improve recovery rates, ensure safety, and minimize secondary pollution. In particular, technological improvement in processing spent batteries is necessary for industrial applications. Therefore, some mandatory improvements do not appear to be within the recycling procedures but are more likely in the design-for-recycling approach, which must be supported.

7. Conclusions

As the demand for lithium-ion batteries continues to grow, there is an increasing need to recover and recycle spent LIBs. This is due to the potential environmental and health risks associated with battery waste, which can impact society's sustainable development. At the same time, spent LIBs represent a valuable resource for future battery materials. This paper provides an overview of current recycling technologies while emphasizing the challenges associated with developing LIB recycling.

One of the major challenges is the need for technological advancements to address the complexities of LIBs, including the various chemistries and designs. Additionally, there are policy gaps that hinder the development of an efficient and effective recycling system. Balancing the economic benefits of recycling with the environmental impact of spent LIBs is also a significant challenge to provide more comprehensive and accurate assessments of the environmental impacts of batteries. However, there are opportunities arising from these challenges, such as the potential for innovation and the creation of a more sustainable and circular economy.

To address end-of-life concerns, this is fundamental to the battery's design. Manufacturing processes that produce cells and packs that are easier to recycle are required. In this context, standardizing cell construction, including cell module design and mechanisms, can help mitigate recycling challenges. The use of intelligent robots can also improve production efficiency and safety by replacing human workers in performing hazardous tasks.

To reduce economic and environmental costs, enhancing the economic efficiency of LIB recycling is crucial. This can be achieved through reducing production costs, increasing product value, on-site recycling, minimizing transportation costs, integrating secondary use with recycling, mitigating negative impacts of primary material extraction and LIB waste, minimizing heavy metal content reduction, and selecting environmentally friendly binders and electrolyte systems.

A comprehensive strategy for battery recycling, known as "design for recycling", is required to optimize both the batteries and the recycling processes simultaneously. This process extends from the physical design and assembly of batteries to simplify recycling, increase yields, and optimize materials, thereby supporting cost-effective battery recycling processes.

The recycling of LIBs contributes to achieving several United Nations Sustainable Development Goals (SDGs). Firstly, SDG 7 (Affordable and Clean Energy) can be supported through LIBs recycling because LIBs are used in energy storage applications, including EVs and renewable energy systems. By recycling spent LIBs, valuable metals can be recovered and reused, reducing the need for new raw materials and promoting a more sustainable approach to energy storage. Secondly, SDG 12 (Responsible Consumption and Production) is connected to LIB recycling because it helps to reduce waste and promote a circular economy. By recovering valuable materials from spent batteries, the need for new resource extraction can be minimized, leading to more responsible consumption and production practices. Thirdly, SDG 13 (Climate Action) can also be advanced by these activities. The production of LIBs involves the extraction and processing of raw materials, which can have significant environmental impacts. It is evident that the battery is mandatory for electric vehicle production, with evident positive consequences in the reduction of greenhouse gases. Moreover, by recycling spent LIBs, the environmental footprint of battery production can be reduced, contributing to climate change mitigation efforts. Furthermore, SDG 14 (Life Below Water) and SDG 15 (Life on Land) can also be supported through recycling because improper disposal of spent batteries can lead to the release of hazardous substances into the environment, posing risks to aquatic and terrestrial ecosystems. By recycling LIBs, the potential for environmental contamination can be minimized, protecting both marine and terrestrial life.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/batteries10010038/s1, Table S1: Scopus results LIBs recovery.

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References

1. IEA. *Global EV Outlook 2020: Entering the Decade of Electric Drive?* Global EV Outlook; OECD Publishing: Paris, France, 2021; ISBN 9789264616226.
2. Miao, Y.; Liu, L.; Zhang, Y.; Tan, Q.; Li, J. An Overview of Global Power Lithium-Ion Batteries and Associated Critical Metal Recycling. *J. Hazard. Mater.* **2022**, *425*, 127900. [CrossRef] [PubMed]
3. Insinna, T.; Bassey, E.N.; Märker, K.; Collauto, A.; Barra, A.L.; Grey, C.P. Graphite Anodes for Li-Ion Batteries: An Electron Paramagnetic Resonance Investigation. *Chem. Mater.* **2023**, *35*, 5497–5511. [CrossRef] [PubMed]
4. Abaza, A.; Ferrari, S.; Wong, H.K.; Lyness, C.; Moore, A.; Weaving, J.; Blanco-Martin, M.; Dashwood, R.; Bhagat, R. Experimental Study of Internal and External Short Circuits of Commercial Automotive Pouch Lithium-Ion Cells. *J. Energy Storage* **2018**, *16*, 211–217. [CrossRef]
5. Goodenough, J.B.; Park, K.S. The Li-Ion Rechargeable Battery: A Perspective. *J. Am. Chem. Soc.* **2013**, *135*, 1167–1176. [CrossRef]
6. 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. [CrossRef]
7. Ding, Y.; Cano, Z.P.; Yu, A.; Lu, J.; Chen, Z. Automotive Li-Ion Batteries: Current Status and Future Perspectives. *Electrochem. Energy Rev.* **2019**, *2*, 1–28. [CrossRef]
8. Ferrara, C.; Ruffo, R.; Quartarone, E.; Mustarelli, P. Circular Economy and the Fate of Lithium Batteries: Second Life and Recycling. *Adv. Energy Sustain. Res.* **2021**, *2*, 2100047. [CrossRef]
9. IEA. *Global EV Outlook 2021: Accelerating Ambitions Despite the Pandemic*; Global EV Outlook; OECD Publishing: Paris, France, 2020; ISBN 9789264723863.
10. Greim, P.; Solomon, A.A.; Breyer, C. Assessment of Lithium Criticality in the Global Energy Transition and Addressing Policy Gaps in Transportation. *Nat. Commun.* **2020**, *11*, 4570. [CrossRef]
11. Marinaro, M.; Bresser, D.; Beyer, E.; Faguy, P.; Hosoi, K.; Li, H.; Sakovica, J.; Amine, K.; Wohlfahrt-Mehrens, M.; Passerini, S.; et al. Bringing Forward the Development of Battery Cells for Automotive Applications: Perspective of R&D Activities in China, Japan, the EU and the USA. *J. Power Sources* **2020**, *459*, 228073. [CrossRef]
12. The Roadmap—Battery 2030+. 2020. Available online: https://battery2030.eu/research/roadmap/ (accessed on 29 December 2023).

13. Amnesty International. *"This Is. What We Die for": Human Rights Abuses in the Democratic Republic of the Congo Power the Global Trade in Cobalt*; Amnesty International: London, UK, 2016.
14. Scheele, F.; De Haan, E.; Kiezebrink, V. Cobalt Blues Environmental Pollution and Human Rights Violations in Katanga's Copper and Cobalt Mines. 2016. Available online: https://www.somo.nl/nl/wp-content/uploads/sites/2/2016/04/Cobalt-blues.pdf (accessed on 17 January 2024).
15. Sommer, P.; Rotter, V.S.; Ueberschaar, M. Battery Related Cobalt and REE Flows in WEEE Treatment. *Waste Manag.* **2015**, *45*, 298–305. [CrossRef]
16. Sverdrup, H.U. Modelling Global Extraction, Supply, Price and Depletion of the Extractable Geological Resources with the LITHIUM Model. *Resour. Conserv. Recycl.* **2016**, *114*, 112–129. [CrossRef]
17. McKinsey & Company. *The Race to Decarbonize Electric-Vehicle Batteries*; McKinsey & Company: New York, NY, USA, 2023.
18. Xu, P.; Tan, D.H.S.; Jiao, B.; Gao, H.; Yu, X.; Chen, Z. A Materials Perspective on Direct Recycling of Lithium-Ion Batteries: Principles, Challenges and Opportunities. *Adv. Funct. Mater.* **2023**, *33*, 2213168. [CrossRef]
19. Wang, J.; Zhang, Q.; Sheng, J.; Liang, Z.; Ma, J.; Chen, Y.; Zhou, G.; Cheng, H.-M. Direct and Green Repairing of Degraded $LiCoO_2$ for Reuse in Lithium-Ion Batteries. *Natl. Sci. Rev.* **2022**, *9*, nwac097. [CrossRef] [PubMed]
20. Sloop, S.E.; Crandon, L.; Allen, M.; Lerner, M.M.; Zhang, H.; Sirisaksoontorn, W.; Gaines, L.; Kim, J.; Lee, M. Cathode Healing Methods for Recycling of Lithium-Ion Batteries. *Sustain. Mater. Technol.* **2019**, *22*, e00113. [CrossRef]
21. Wan, J.; Lyu, J.; Bi, W.; Zhou, Q.; Li, P.; Li, H.; Li, Y. Regeneration of Spent Lithium-Ion Battery Materials. *J. Energy Storage* **2022**, *51*, 104470. [CrossRef]
22. Anwani, S.; Methekar, R.; Ramadesigan, V. Resynthesizing of Lithium Cobalt Oxide from Spent Lithium-Ion Batteries Using an Environmentally Benign and Economically Viable Recycling Process. *Hydrometallurgy* **2020**, *197*, 105430. [CrossRef]
23. Li, L.; Chen, R.; Sun, F.; Wu, F.; Liu, J. Preparation of $LiCoO_2$ Films from Spent Lithium-Ion Batteries by a Combined Recycling Process. *Hydrometallurgy* **2011**, *108*, 220–225. [CrossRef]
24. Zhang, X.; Xie, Y.; Lin, X.; Li, H.; Cao, H. An Overview on the Processes and Technologies for Recycling Cathodic Active Materials from Spent Lithium-Ion Batteries. *J. Mater. Cycles Waste Manag.* **2013**, *15*, 420–430. [CrossRef]
25. Zhang, X.; Xie, Y.; Cao, H.; Nawaz, F.; Zhang, Y. A Novel Process for Recycling and Resynthesizing $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ from the Cathode Scraps Intended for Lithium-Ion Batteries. *Waste Manag.* **2014**, *34*, 1715–1724. [CrossRef]
26. Natarajan, S.; Aravindan, V. Burgeoning Prospects of Spent Lithium-Ion Batteries in Multifarious Applications. *Adv. Energy Mater.* **2018**, *8*, 1802303. [CrossRef]
27. 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]
28. Khodadadmahmoudi, G.; Javdan Tabar, K.; Homayouni, A.H.; Chehreh Chelgani, S. Recycling Spent Lithium Batteries—An Overview of Pretreatment Flowsheet Development Based on Metallurgical Factors. *Environ. Technol. Rev.* **2023**, *12*, 2248559. [CrossRef]
29. Werner, D.M.; Mütze, T.; Peuker, U.A. Influence of Cell Opening Methods on Organic Solvent Removal during Pretreatment in Lithium-Ion Battery Recycling. *Waste Manag. Res.* **2022**, *40*, 1015–1026. [CrossRef]
30. 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]
31. 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]
32. Yu, D.; Huang, Z.; Makuza, B.; Guo, X.; Tian, Q. Pretreatment Options for the Recycling of Spent Lithium-Ion Batteries: A Comprehensive Review. *Miner. Eng.* **2021**, *173*, 107218. [CrossRef]
33. Yu, W.; Guo, Y.; Xu, S.; Yang, Y.; Zhao, Y.; Zhang, J. Comprehensive Recycling of Lithium-Ion Batteries: Fundamentals, Pretreatment, and Perspectives. *Energy Storage Mater.* **2023**, *54*, 172–220. [CrossRef]
34. Liu, C.; Lin, J.; Cao, H.; Zhang, Y.; Sun, Z. Recycling of Spent Lithium-Ion Batteries in View of Lithium Recovery: A Critical Review. *J. Clean. Prod.* **2019**, *228*, 801–813. [CrossRef]
35. Amalia, D.; Singh, P.; Zhang, W.; Nikoloski, A.N. Discharging of Spent Cylindrical Lithium-Ion Batteries in Sodium Hydroxide and Sodium Chloride for a Safe Recycling Process. *JOM* **2023**, *75*, 4946–4957. [CrossRef]
36. Tian, G.; Yuan, G.; Aleksandrov, A.; Zhang, T.; Li, Z.; Fathollahi-Fard, A.M.; Ivanov, M. Recycling of Spent Lithium-Ion Batteries: A Comprehensive Review for Identification of Main Challenges and Future Research Trends. *Sustain. Energy Technol. Assess.* **2022**, *53*, 102447. [CrossRef]
37. Wu, S.; Kaden, N.; Dröder, K. A Systematic Review on Lithium-Ion Battery Disassembly Processes for Efficient Recycling. *Batteries* **2023**, *9*, 297. [CrossRef]
38. Meng, K.; Xu, G.; Peng, X.; Youcef-Toumi, K.; Li, J. Intelligent Disassembly of Electric-Vehicle Batteries: A Forward-Looking Overview. *Resour. Conserv. Recycl.* **2022**, *182*, 106207. [CrossRef]
39. Lin, J.; Wu, J.; Fan, E.; Zhang, X.; Chen, R.; Wu, F.; Li, L. Environmental and Economic Assessment of Structural Repair Technologies for Spent Lithium-Ion Battery Cathode Materials. *Int. J. Miner. Metall. Mater.* **2022**, *29*, 942–952. [CrossRef]
40. 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]

41. 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]
42. Yang, T.; Luo, D.; Yu, A.; Chen, Z. Enabling Future Closed-Loop Recycling of Spent Lithium-Ion Batteries: Direct Cathode Regeneration. *Adv. Mater.* **2023**, *35*, 2203218. [CrossRef]
43. Zhang, R.; Shi, X.; Esan, O.C.; An, L. Organic Electrolytes Recycling From Spent Lithium-Ion Batteries. *Glob. Chall.* **2022**, *6*, 2200050. [CrossRef]
44. Shi, G.; Cheng, J.; Wang, J.; Zhang, S.; Shao, X.; Chen, X.; Li, X.; Xin, B. A Comprehensive Review of Full Recycling and Utilization of Cathode and Anode as Well as Electrolyte from Spent Lithium-Ion Batteries. *J. Energy Storage* **2023**, *72*, 108486. [CrossRef]
45. Qiao, Y.; Zhao, H.; Shen, Y.; Li, L.; Rao, Z.; Shao, G.; Lei, Y. Recycling of Graphite Anode from Spent Lithium-ion Batteries: Advances and Perspectives. *EcoMat* **2023**, *5*, e12321. [CrossRef]
46. Niu, B.; Xiao, J.; Xu, Z. Advances and Challenges in Anode Graphite Recycling from Spent Lithium-Ion Batteries. *J. Hazard. Mater.* **2022**, *439*, 129678. [CrossRef]
47. 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]
48. Maisel, F.; Neef, C.; Marscheider-Weidemann, F.; Nissen, N.F. A Forecast on Future Raw Material Demand and Recycling Potential of Lithium-Ion Batteries in Electric Vehicles. *Resour. Conserv. Recycl.* **2023**, *192*, 106920. [CrossRef]
49. Huang, B.; Pan, Z.; Su, X.; An, L. Recycling of Lithium-Ion Batteries: Recent Advances and Perspectives. *J. Power Sources* **2018**, *399*, 274–286. [CrossRef]
50. Latini, D.; Vaccari, M.; Lagnoni, M.; Orefice, M.; Mathieux, F.; Huisman, J.; Tognotti, L.; Bertei, A. A Comprehensive Review and Classification of Unit Operations with Assessment of Outputs Quality in Lithium-Ion Battery Recycling. *J. Power Sources* **2022**, *546*, 231979. [CrossRef]
51. Golmohammadzadeh, R.; Faraji, F.; Jong, B.; Pozo-Gonzalo, C.; Banerjee, P.C. Current Challenges and Future Opportunities toward Recycling of Spent Lithium-Ion Batteries. *Renew. Sustain. Energy Rev.* **2022**, *159*, 112202. [CrossRef]
52. Sheth, R.P.; Ranawat, N.S.; Chakraborty, A.; Mishra, R.P.; Khandelwal, M. The Lithium-Ion Battery Recycling Process from a Circular Economy Perspective—A Review and Future Directions. *Energies* **2023**, *16*, 3228. [CrossRef]
53. 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.* **2021**, *55*, 391–419. [CrossRef]
54. Statista Value of Recycled EV Batteries by Battery Type. Available online: https://www.statista.com/statistics/1246828/value-of-recycled-ev-batteries-by-battery-type/ (accessed on 29 September 2023).
55. van Halm, I.; Mullan, C. Booming EV Sales Challenge Critical Mineral Supply Chains. Available online: https://www.energymonitor.ai/sectors/transport/booming-ev-sales-challenge-mineral-supply-chains/ (accessed on 17 January 2024).
56. Widijatmoko, S.D.; Fu, G.; Wang, Z.; Hall, P. Recovering Lithium Cobalt Oxide, Aluminium, and Copper from Spent Lithium-Ion Battery via Attrition Scrubbing. *J. Clean. Prod.* **2020**, *260*, 120869. [CrossRef]
57. Widijatmoko, S.D.; Gu, F.; Wang, Z.; Hall, P. Selective Liberation in Dry Milled Spent Lithium-Ion Batteries. *Sustain. Mater. Technol.* **2020**, *23*, e00134. [CrossRef]
58. Mossali, E.; Picone, N.; Gentilini, L.; Rodrìguez, O.; Pérez, J.M.; Colledani, M. Lithium-Ion Batteries towards Circular Economy: A Literature Review of Opportunities and Issues of Recycling Treatments. *J. Environ. Manag.* **2020**, *264*, 110500. [CrossRef]
59. Yu, W.; Zhang, Y.; Hu, J.; Zhou, J.; Shang, Z.; Zhou, X.; Xu, S. Controlled Carbothermic Reduction for Enhanced Recovery of Metals from Spent Lithium-Ion Batteries. *Resour. Conserv. Recycl.* **2023**, *194*, 107005. [CrossRef]
60. Makuza, B.; Yu, D.; Huang, Z.; Guo, X.; Tian, Q.; Zhang, K.; Zhang, B.; Liu, P. Synergetic Carbothermic Reduction and Selective Hydrochlorination of Spent Li-Ion Batteries Black Mass towards Enhanced Metal Recovery. *J. Clean. Prod.* **2023**, *386*, 135831. [CrossRef]
61. Li, J.; Li, L.; Yang, R.; Jiao, J. Assessment of the Lifecycle Carbon Emission and Energy Consumption of Lithium-Ion Power Batteries Recycling: A Systematic Review and Meta-Analysis. *J. Energy Storage* **2023**, *65*, 107306. [CrossRef]
62. Omran, M.; Fabritius, T.; Heikkinen, E.-P.; Vuolio, T.; Yu, Y.; Chen, G.; Kacar, Y. Microwave Catalyzed Carbothermic Reduction of Zinc Oxide and Zinc Ferrite: Effect of Microwave Energy on the Reaction Activation Energy. *RSC Adv.* **2020**, *10*, 23959–23968. [CrossRef]
63. Fahimi, A.; Alessandri, I.; Cornelio, A.; Frontera, P.; Malara, A.; Mousa, E.; Ye, G.; Valentim, B.; Bontempi, E. A Microwave-Enhanced Method Able to Substitute Traditional Pyrometallurgy for the Future of Metals Supply from Spent Lithium-Ion Batteries. *Resour. Conserv. Recycl.* **2023**, *194*, 106989. [CrossRef]
64. Fan, X.; Song, C.; Lu, X.; Shi, Y.; Yang, S.; Zheng, F.; Huang, Y.; Liu, K.; Wang, H.; Li, Q. Separation and Recovery of Valuable Metals from Spent Lithium-Ion Batteries via Concentrated Sulfuric Acid Leaching and Regeneration of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$. *J. Alloys Compd.* **2021**, *863*, 158775. [CrossRef]
65. Zheng, X.; Gao, W.; Zhang, X.; He, M.; Lin, X.; Cao, H.; Zhang, Y.; Sun, Z. Spent Lithium-Ion Battery Recycling-Reductive Ammonia Leaching of Metals from Cathode Scrap by Sodium Sulphite. *Waste Manag.* **2016**, *60*, 680–688. [CrossRef]
66. 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]

67. Wang, H.; Huang, K.; Zhang, Y.; Chen, X.; Jin, W.; Zheng, S.; Zhang, Y.; Li, P. Recovery of Lithium, Nickel, and Cobalt from Spent Lithium-Ion Battery Powders by Selective Ammonia Leaching and an Adsorption Separation System. *ACS Sustain. Chem. Eng.* **2017**, *5*, 11489–11495. [CrossRef]
68. Gaye, N.; Gueye, R.S.; Ledauphin, J.; Balde, M.; Seck, M.; Wele, A.; Diaw, M. Alkaline Leaching of Metals from Cathodic Materials of Spent Lithium-Ion Batteries. *Asian J. Appl. Chem. Res.* **2019**, *3*, 1–7. [CrossRef]
69. Biswal, B.K.; Balasubramanian, R. Recovery of Valuable Metals from Spent Lithium-Ion Batteries Using Microbial Agents for Bioleaching: A Review. *Front. Microbiol.* **2023**, *14*, 1197081. [CrossRef]
70. 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]
71. Kumari, A.; Jha, M.K.; Lee, J.; Singh, R.P. Clean Process for Recovery of Metals and Recycling of Acid from the Leach Liquor of PCBs. *J. Clean. Prod.* **2016**, *112*, 4826–4834. [CrossRef]
72. Ning, P.; Meng, Q.; Dong, P.; Duan, J.; Xu, M.; Lin, Y.; Zhang, Y. Recycling of Cathode Material from Spent Lithium Ion Batteries Using an Ultrasound-Assisted DL-Malic Acid Leaching System. *Waste Manag.* **2020**, *103*, 52–60. [CrossRef] [PubMed]
73. Zeng, X.; Li, J.; Shen, B. Novel Approach to Recover Cobalt and Lithium from Spent Lithium-Ion Battery Using Oxalic Acid. *J. Hazard. Mater.* **2015**, *295*, 112–118. [CrossRef]
74. Li, L.; Ge, J.; Wu, F.; Chen, R.; Chen, S.; Wu, B. Recovery of Cobalt and Lithium from Spent Lithium Ion Batteries Using Organic Citric Acid as Leachant. *J. Hazard. Mater.* **2010**, *176*, 288–293. [CrossRef] [PubMed]
75. Nayaka, G.P.; Manjanna, J.; Pai, K.V.; Vadavi, R.; Keny, S.J.; Tripathi, V.S. Recovery of Valuable Metal Ions from the Spent Lithium-Ion Battery Using Aqueous Mixture of Mild Organic Acids as Alternative to Mineral Acids. *Hydrometallurgy* **2015**, *151*, 73–77. [CrossRef]
76. Zeng, G.; Yao, J.; Liu, C.; Luo, X.; Ji, H.; Mi, X.; Deng, C. Simultaneous Recycling of Critical Metals and Aluminum Foil from Waste $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ Cathode via Ethylene Glycol–Citric Acid System. *ACS Sustain. Chem. Eng.* **2021**, *9*, 16133–16142. [CrossRef]
77. Moazzam, P.; Boroumand, Y.; Rabiei, P.; Baghbaderani, S.S.; Mokarian, P.; Mohagheghian, F.; Mohammed, L.J.; Razmjou, A. Lithium Bioleaching: An Emerging Approach for the Recovery of Li from Spent Lithium Ion Batteries. *Chemosphere* **2021**, *277*, 130196. [CrossRef]
78. Zhang, W.; Xu, C.; He, W.; Li, G.; Huang, J. A Review on Management of Spent Lithium Ion Batteries and Strategy for Resource Recycling of All Components from Them. *Waste Manag. Res. J. A Sustain. Circ. Econ.* **2018**, *36*, 99–112. [CrossRef]
79. Bahaloo-Horeh, N.; Mousavi, S.M. Enhanced Recovery of Valuable Metals from Spent Lithium-Ion Batteries through Optimization of Organic Acids Produced by Aspergillus Niger. *Waste Manag.* **2017**, *60*, 666–679. [CrossRef]
80. Esmaeili, M.; Rastegar, S.O.; Beigzadeh, R.; Gu, T. Ultrasound-Assisted Leaching of Spent Lithium Ion Batteries by Natural Organic Acids and H_2O_2. *Chemosphere* **2020**, *254*, 126670. [CrossRef] [PubMed]
81. Li, B.; Li, Q.; Wang, Q.; Yan, X.; Shi, M.; Wu, C. Deep Eutectic Solvent for Spent Lithium-Ion Battery Recycling: Comparison with Inorganic Acid Leaching. *Phys. Chem. Chem. Phys.* **2022**, *24*, 19029–19051. [CrossRef] [PubMed]
82. Golmohammadzadeh, R.; Faraji, F.; Rashchi, F. Recovery of Lithium and Cobalt from Spent Lithium Ion Batteries (LIBs) Using Organic Acids as Leaching Reagents: A Review. *Resour. Conserv. Recycl.* **2018**, *136*, 418–435. [CrossRef]
83. de Oliveira Demarco, J.; Stefanello Cadore, J.; da Silveira de Oliveira, F.; Hiromitsu Tanabe, E.; Assumpção Bertuol, D. Recovery of Metals from Spent Lithium-Ion Batteries Using Organic Acids. *Hydrometallurgy* **2019**, *190*, 105169. [CrossRef]
84. Wang, S.; Zhang, Z.; Lu, Z.; Xu, Z. A Novel Method for Screening Deep Eutectic Solvent to Recycle the Cathode of Li-Ion Batteries. *Green Chem.* **2020**, *22*, 4473–4482. [CrossRef]
85. Wang, M.; Liu, K.; Xu, Z.; Dutta, S.; Valix, M.; Alessi, D.S.; Huang, L.; Zimmerman, J.B.; Tsang, D.C.W. Selective Extraction of Critical Metals from Spent Lithium-Ion Batteries. *Environ. Sci. Technol.* **2023**, *57*, 3940–3950. [CrossRef]
86. Suriyanarayanan, S.; Babu, M.P.; Murugan, R.; Muthuraj, D.; Ramanujam, K.; Nicholls, I.A. Highly Efficient Recovery and Recycling of Cobalt from Spent Lithium-Ion Batteries Using an *N*-Methylurea–Acetamide Nonionic Deep Eutectic Solvent. *ACS Omega* **2023**, *8*, 6959–6967. [CrossRef]
87. Martín, M.I.; García-Díaz, I.; López, F.A. Properties and Perspective of Using Deep Eutectic Solvents for Hydrometallurgy Metal Recovery. *Miner. Eng.* **2023**, *203*, 108306. [CrossRef]
88. Chen, Y.; Lu, Y.; Liu, Z.; Zhou, L.; Li, Z.; Jiang, J.; Wei, L.; Ren, P.; Mu, T. Efficient Dissolution of Lithium-Ion Batteries Cathode $LiCoO_2$ by Polyethylene Glycol-Based Deep Eutectic Solvents at Mild Temperature. *ACS Sustain. Chem. Eng.* **2020**, *8*, 11713–11720. [CrossRef]
89. He, X.; Wen, Y.; Wang, X.; Cui, Y.; Li, L.; Ma, H. Leaching NCM Cathode Materials of Spent Lithium-Ion Batteries with Phosphate Acid-Based Deep Eutectic Solvent. *Waste Manag.* **2023**, *157*, 8–16. [CrossRef]
90. Huang, F.; Li, T.; Yan, X.; Xiong, Y.; Zhang, X.; Lu, S.; An, N.; Huang, W.; Guo, Q.; Ge, X. Ternary Deep Eutectic Solvent (DES) with a Regulated Rate-Determining Step for Efficient Recycling of Lithium Cobalt Oxide. *ACS Omega* **2022**, *7*, 11452–11459. [CrossRef]
91. Wang, M.; Tan, Q.; Liu, L.; Li, J. A Low-Toxicity and High-Efficiency Deep Eutectic Solvent for the Separation of Aluminum Foil and Cathode Materials from Spent Lithium-Ion Batteries. *J. Hazard. Mater.* **2019**, *380*, 120846. [CrossRef]

92. Yu, H.; Wang, S.; Li, Y.; Qiao, Q.; Wang, K.; Li, X. Recovery of Cobalt from Spent Lithium-Ion Battery Cathode Materials by Using Choline Chloride-Based Deep Eutectic Solvent. *Green Process. Synth.* **2022**, *11*, 868–874. [CrossRef]
93. Padwal, C.; Pham, H.D.; Jadhav, S.; Do, T.T.; Nerkar, J.; Hoang, L.T.M.; Kumar Nanjundan, A.; Mundree, S.G.; Dubal, D.P. Deep Eutectic Solvents: Green Approach for Cathode Recycling of Li-Ion Batteries. *Adv. Energy Sustain. Res.* **2022**, *3*, 2100133. [CrossRef]
94. Wang, Z.; Li, S.; Li, T.; Hu, T.; Ge, X. Deep Eutectic Solvents (DESs) for Green Recycling of Wasted Lithium-Ion Batteries (LIBs): Progress on Pushing the Overall Efficiency. *Min. Metall. Explor.* **2022**, *39*, 2149–2165. [CrossRef]
95. Wang, K.; Hu, T.; Shi, P.; Min, Y.; Wu, J.; Xu, Q. Efficient Recovery of Value Metals from Spent Lithium-Ion Batteries by Combining Deep Eutectic Solvents and Coextraction. *ACS Sustain. Chem. Eng.* **2022**, *10*, 1149–1159. [CrossRef]
96. Wang, H.; Li, M.; Garg, S.; Wu, Y.; Nazmi Idros, M.; Hocking, R.; Duan, H.; Gao, S.; Yago, A.J.; Zhuang, L.; et al. Cobalt Electrochemical Recovery from Lithium Cobalt Oxides in Deep Eutectic Choline Chloride+Urea Solvents. *ChemSusChem* **2021**, *14*, 2972–2983. [CrossRef]
97. Tran, M.K.; Rodrigues, M.-T.F.; Kato, K.; Babu, G.; Ajayan, P.M. Deep Eutectic Solvents for Cathode Recycling of Li-Ion Batteries. *Nat. Energy* **2019**, *4*, 339–345. [CrossRef]
98. Tang, S.; Zhang, M.; Guo, M. A Novel Deep-Eutectic Solvent with Strong Coordination Ability and Low Viscosity for Efficient Extraction of Valuable Metals from Spent Lithium-Ion Batteries. *ACS Sustain. Chem. Eng.* **2022**, *10*, 975–985. [CrossRef]
99. Chang, X.; Fan, M.; Gu, C.; He, W.; Meng, Q.; Wan, L.; Guo, Y. Selective Extraction of Transition Metals from Spent $LiNi_xCo_y Mn_{1-x-y}O_2$ Cathode via Regulation of Coordination Environment. *Angew. Chem. Int. Ed.* **2022**, *61*, e202202558. [CrossRef] [PubMed]
100. Ma, C.; Svärd, M.; Forsberg, K. Recycling Cathode Material $LiCo_{1/3}Ni_{1/3}Mn_{1/3}O_2$ by Leaching with a Deep Eutectic Solvent and Metal Recovery with Antisolvent Crystallization. *Resour. Conserv. Recycl.* **2022**, *186*, 106579. [CrossRef]
101. Lai, Y.; Zhu, X.; Li, J.; Gou, Q.; Li, M.; Xia, A.; Huang, Y.; Zhu, X.; Liao, Q. Recovery and Regeneration of Anode Graphite from Spent Lithium-Ion Batteries through Deep Eutectic Solvent Treatment: Structural Characteristics, Electrochemical Performance and Regeneration Mechanism. *Chem. Eng. J.* **2023**, *457*, 141196. [CrossRef]
102. Hua, Y.; Xu, Z.; Zhao, B.; Zhang, Z. Efficient Separation of Electrode Active Materials and Current Collector Metal Foils from Spent Lithium-Ion Batteries by a Green Deep Eutectic Solvent. *Green Chem.* **2022**, *24*, 8131–8141. [CrossRef]
103. Gong, S.; Wu, T.; Wang, G.; Huang, Q.; Su, Y.; Wu, F. Screening of Deep Eutectic Solvent Based on Efficient Recovery of Spent Lithium-Ion Battery Cathode Materials. *Gaodeng Xuexiao Huaxue Xuebao/Chem. J. Chin. Univ.* **2021**, *42*, 3151–3159. [CrossRef]
104. Council of the European Union; European Parliament. *European Commission Regulation (EU) 2023/1542 of the European Parliament and of the Council Concerning Batteries and Waste Batteries*; European Commission: Brussels, Belgium, 2023; Volume 2023, pp. 1–117.
105. Gulliani, S.; Volpe, M.; Messineo, A.; Volpe, R. Recovery of Metals and Valuable Chemicals from Waste Electric and Electronic Materials: A Critical Review of Existing Technologies. *RSC Sustain.* **2023**, *1*, 1085–1108. [CrossRef]
106. Ma, J.; Wang, J.; Jia, K.; Liang, Z.; Ji, G.; Zhuang, Z.; Zhou, G.; Cheng, H.-M. Adaptable Eutectic Salt for the Direct Recycling of Highly Degraded Layer Cathodes. *J. Am. Chem. Soc.* **2022**, *144*, 20306–20314. [CrossRef]
107. Graham, J.D.; Rupp, J.A.; Brungard, E. Lithium in the Green Energy Transition: The Quest for Both Sustainability and Security. *Sustainability* **2021**, *13*, 11274. [CrossRef]
108. Fan, X.; Tan, C.; Li, Y.; Chen, Z.; Li, Y.; Huang, Y.; Pan, Q.; Zheng, F.; Wang, H.; Li, Q. A Green, Efficient, Closed-Loop Direct Regeneration Technology for Reconstructing of the $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ Cathode Material from Spent Lithium-Ion Batteries. *J. Hazard. Mater.* **2021**, *410*, 124610. [CrossRef]
109. Natarajan, S.; Aravindan, V. An Urgent Call to Spent LIB Recycling: Whys and Wherefores for Graphite Recovery. *Adv. Energy Mater.* **2020**, *10*, 2002238. [CrossRef]
110. Zhang, G.; He, Y.; Wang, H.; Feng, Y.; Xie, W.; Zhu, X. Application of Mechanical Crushing Combined with Pyrolysis-Enhanced Flotation Technology to Recover Graphite and $LiCoO_2$ from Spent Lithium-Ion Batteries. *J. Clean. Prod.* **2019**, *231*, 1418–1427. [CrossRef]
111. Xu, R.; Lei, S.; Wang, T.; Yi, C.; Sun, W.; Yang, Y. Lithium Recovery and Solvent Reuse from Electrolyte of Spent Lithium-Ion Battery. *Waste Manag.* **2023**, *167*, 135–140. [CrossRef]
112. Fu, Y.; Schuster, J.; Petranikova, M.; Ebin, B. Innovative Recycling of Organic Binders from Electric Vehicle Lithium-Ion Batteries by Supercritical Carbon Dioxide Extraction. *Resour. Conserv. Recycl.* **2021**, *172*, 105666. [CrossRef]
113. Jung, S.; Kwon, D.; Park, S.; Kwon, K.; Tsang, Y.F.; Kwon, E.E. Valorization of a Spent Lithium-Ion Battery Electrolyte through Syngas Formation Using CO_2-Assisted Catalytic Thermolysis over a Battery Cathode Material. *J. CO_2 Util.* **2021**, *50*, 101591. [CrossRef]
114. Natarajan, S.; Akshay, M.; Aravindan, V. Recycling/Reuse of Current Collectors from Spent Lithium-Ion Batteries: Benefits and Issues. *Adv. Sustain. Syst.* **2022**, *6*, 2100432. [CrossRef]
115. Zhong, X.; Liu, W.; Han, J.; Jiao, F.; Qin, W.; Liu, T.; Zhao, C. Pyrolysis and Physical Separation for the Recovery of Spent $LiFePO_4$ Batteries. *Waste Manag.* **2019**, *89*, 83–93. [CrossRef] [PubMed]
116. Wu, Z.; Zhu, H.; Bi, H.; He, P.; Gao, S. Recycling of Electrode Materials from Spent Lithium-Ion Power Batteries via Thermal and Mechanical Treatments. *Waste Manag. Res.* **2021**, *39*, 607–619. [CrossRef] [PubMed]
117. Chen, X.; Li, S.; Wang, Y.; Jiang, Y.; Tan, X.; Han, W.; Wang, S. Recycling of $LiFePO_4$ Cathode Materials from Spent Lithium-Ion Batteries through Ultrasound-Assisted Fenton Reaction and Lithium Compensation. *Waste Manag.* **2021**, *136*, 67–75. [CrossRef]

118. Bai, Y.; Belharouak, I. Solvent-Based Electrode Recovery Toward Sustainable Direct Recycling of Lithium-Ion Batteries. *ECS Meet. Abstr.* **2022**, *MA2022-02*, 341. [CrossRef]
119. Paone, E.; Miceli, M.; Malara, A.; Ye, G.; Mousa, E.; Bontempi, E.; Frontera, P.; Mauriello, F. Direct Reuse of Spent Lithium-Ion Batteries as an Efficient Heterogeneous Catalyst for the Reductive Upgrading of Biomass-Derived Furfural. *ACS Sustain. Chem. Eng.* **2022**, *10*, 2275–2281. [CrossRef]
120. Ducoli, S.; Fahimi, A.; Mousa, E.; Ye, G.; Federici, S.; Frontera, P.; Bontempi, E. ESCAPE Approach for the Sustainability Evaluation of Spent Lithium-Ion Batteries Recovery: Dataset of 33 Available Technologies. *Data Brief.* **2022**, *42*, 108018. [CrossRef]
121. Fahimi, A.; Zanoletti, A.; Cornelio, A.; Mousa, E.; Ye, G.; Frontera, P.; Depero, L.E.; Bontempi, E. Sustainability Analysis of Processes to Recycle Discharged Lithium-Ion Batteries, Based on the ESCAPE Approach. *Materials* **2022**, *15*, 8527. [CrossRef] [PubMed]
122. Bontempi, E. How to Perform a Material Recovery Sustainability Evaluation Preliminary to LCA? *Resour. Environ. Sustain.* **2022**, *9*, 100074. [CrossRef]
123. Dorrmann, L.; Sann-Ferro, K.; Heininger, P.; Mähliß, J. *Kompendium: Li-Ionen-Batterien*; VDE Studie: Offenbach, Germany, 2021.
124. Sandaka, B.P.; Kumar, J. Alternative Vehicular Fuels for Environmental Decarbonization: A Critical Review of Challenges in Using Electricity, Hydrogen, and Biofuels as a Sustainable Vehicular Fuel. *Chem. Eng. J. Adv.* **2023**, *14*, 100442. [CrossRef]
125. Mousa, E.; Hu, X.; Ånnhagen, L.; Ye, G.; Cornelio, A.; Fahimi, A.; Bontempi, E.; Frontera, P.; Badenhorst, C.; Santos, A.C.; et al. Characterization and Thermal Treatment of the Black Mass from Spent Lithium-Ion Batteries. *Sustainability* **2022**, *15*, 15. [CrossRef]
126. Windisch-Kern, S.; Gerold, E.; Nigl, T.; Jandric, A.; Altendorfer, M.; Rutrecht, B.; Scherhaufer, S.; Raupenstrauch, H.; Pomberger, R.; Antrekowitsch, H.; et al. Recycling Chains for Lithium-Ion Batteries: A Critical Examination of Current Challenges, Opportunities and Process Dependencies. *Waste Manag.* **2022**, *138*, 125–139. [CrossRef] [PubMed]
127. Lima, M.C.C.; Pontes, L.P.; Vasconcelos, A.S.M.; de Araujo Silva Junior, W.; Wu, K. Economic Aspects for Recycling of Used Lithium-Ion Batteries from Electric Vehicles. *Energies* **2022**, *15*, 2203. [CrossRef]
128. Explore 2+ Mio. Startups & Scaleups: StartUs Insights Discovery Platform. Available online: https://www.startus-insights.com/startus-insights-platform/ (accessed on 30 September 2023).
129. Li-Cycle Holdings Corp. *Annual Report*; Li-Cycle Holdings Corp.: Toronto, ON, Canada, 2021.
130. Kallitsis, E.; Korre, A.; Kelsall, G.H. Life Cycle Assessment of Recycling Options for Automotive Li-Ion Battery Packs. *J. Clean. Prod.* **2022**, *371*, 133636. [CrossRef]
131. Majeau-Bettez, G.; Hawkins, T.R.; Strømman, A.H. Life Cycle Environmental Assessment of Lithium-Ion and Nickel Metal Hydride Batteries for Plug-In Hybrid and Battery Electric Vehicles. *Environ. Sci. Technol.* **2011**, *45*, 4548–4554. [CrossRef]
132. Wagner-Wenz, R.; van Zuilichem, A.J.; Göllner-Völker, L.; Berberich, K.; Weidenkaff, A.; Schebek, L. Recycling Routes of Lithium-Ion Batteries: A Critical Review of the Development Status, the Process Performance, and Life-Cycle Environmental Impacts. *MRS Energy Sustain.* **2022**, *10*, 1–34. [CrossRef]
133. Peters, J.F. Best Practices for Life Cycle Assessment of Batteries. *Nat. Sustain.* **2023**, *6*, 614–616. [CrossRef]
134. De Benedetti, B.; Toso, D.; Baldo, G.L.; Rollino, S. EcoAudit: A Renewed Simplified Procedure to Facilitate the Environmentally Informed Material Choice Orienting the Further Life Cycle Analysis for Ecodesigners. *Mater. Trans.* **2010**, *51*, 832–837. [CrossRef]
135. Bontempi, E. A New Approach for Evaluating the Sustainability of Raw Materials Substitution Based on Embodied Energy and the CO_2 Footprint. *J. Clean. Prod.* **2017**, *162*, 162–169. [CrossRef]
136. Bontempi, E. Raw Materials and Sustainability Indicators. In *Raw Materials Substitution Sustainability*; SpringerBriefs in Applied Sciences and Technology; Springer: Cham, Switzerland, 2017; pp. 1–28.
137. Fahimi, A.; Ducoli, S.; Federici, S.; Ye, G.; Mousa, E.; Frontera, P.; Bontempi, E. Evaluation of the Sustainability of Technologies to Recycle Spent Lithium-Ion Batteries, Based on Embodied Energy and Carbon Footprint. *J. Clean. Prod.* **2022**, *338*, 130493. [CrossRef]
138. 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]
139. 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]
140. Neumann, J.; Petranikova, M.; Meeus, M.; Gamarra, J.D.; Younesi, R.; Winter, M.; Nowak, S. Recycling of Lithium-Ion Batteries—Current State of the Art, Circular Economy, and Next Generation Recycling. *Adv. Energy Mater.* **2022**, *12*, 2102917. [CrossRef]
141. Ali, H.; Khan, H.A.; Pecht, M. Preprocessing of Spent Lithium-Ion Batteries for Recycling: Need, Methods, and Trends. *Renew. Sustain. Energy Rev.* **2022**, *168*, 112809. [CrossRef]
142. Xiao, J.; Jiang, C.; Wang, B. A Review on Dynamic Recycling of Electric Vehicle Battery: Disassembly and Echelon Utilization. *Batteries* **2023**, *9*, 57. [CrossRef]
143. Marshall, J.; Gastol, D.; Sommerville, R.; Middleton, B.; Goodship, V.; Kendrick, E. Disassembly of Li Ion Cells—Characterization and Safety Considerations of a Recycling Scheme. *Metals* **2020**, *10*, 773. [CrossRef]
144. Zhou, L.; Garg, A.; Zheng, J.; Gao, L.; Oh, K. Battery Pack Recycling Challenges for the Year 2030: Recommended Solutions Based on Intelligent Robotics for Safe and Efficient Disassembly, Residual Energy Detection, and Secondary Utilization. *Energy Storage* **2021**, *3*, e190. [CrossRef]

145. Alfaro-Algaba, M.; Ramirez, F.J. Techno-Economic and Environmental Disassembly Planning of Lithium-Ion Electric Vehicle Battery Packs for Remanufacturing. *Resour. Conserv. Recycl.* **2020**, *154*, 104461. [CrossRef]
146. Thompson, D.; Hyde, C.; Hartley, J.M.; Abbott, A.P.; Anderson, P.A.; Harper, G.D.J. To Shred or Not to Shred: A Comparative Techno-Economic Assessment of Lithium Ion Battery Hydrometallurgical Recycling Retaining Value and Improving Circularity in LIB Supply Chains. *Resour. Conserv. Recycl.* **2021**, *175*, 105741. [CrossRef]
147. Henriksen, M.; Vaagsaether, K.; Lundberg, J.; Forseth, S.; Bjerketvedt, D. Explosion Characteristics for Li-Ion Battery Electrolytes at Elevated Temperatures. *J. Hazard. Mater.* **2019**, *371*, 1–7. [CrossRef] [PubMed]
148. Lai, X.; Huang, Y.; Deng, C.; Gu, H.; Han, X.; Zheng, Y.; Ouyang, M. Sorting, Regrouping, and Echelon Utilization of the Large-Scale Retired Lithium Batteries: A Critical Review. *Renew. Sustain. Energy Rev.* **2021**, *146*, 111162. [CrossRef]
149. Lee, K.; Kum, D. Development of Cell Selection Framework for Second-Life Cells with Homogeneous Properties. *Int. J. Electr. Power Energy Syst.* **2019**, *105*, 429–439. [CrossRef]
150. Li, X.; Zhang, L.; Liu, Y.; Pan, A.; Liao, Q.; Yang, X. A Fast Classification Method of Retired Electric Vehicle Battery Modules and Their Energy Storage Application in Photovoltaic Generation. *Int. J. Energy Res.* **2020**, *44*, 2337–2344. [CrossRef]
151. Thompson, D.L.; Hartley, J.M.; Lambert, S.M.; Shiref, M.; Harper, G.D.J.; Kendrick, E.; Anderson, P.; Ryder, K.S.; Gaines, L.; Abbott, A.P. The Importance of Design in Lithium Ion Battery Recycling—A Critical Review. *Green Chem.* **2020**, *22*, 7585–7603. [CrossRef]
152. 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]
153. Diekmann, J.; Grutzke, M.; Herrmann, C.; Kara, S.; Kwade, A. *Recycling of Lithium-Ion Batteries*; Kwade, A., Diekmann, J., Eds.; Springer: Berlin/Heidelberg, Germany, 2018; pp. 250–251, ISBN 978-3-319-70571-2.
154. Wang, Q.; Ping, P.; Zhao, X.; Chu, G.; Sun, J.; Chen, C. Thermal Runaway Caused Fire and Explosion of Lithium Ion Battery. *J. Power Sources* **2012**, *208*, 210–224. [CrossRef]
155. Slattery, M.; Dunn, J.; Kendall, A. Transportation of Electric Vehicle Lithium-Ion Batteries at End-of-Life: A Literature Review. *Resour. Conserv. Recycl.* **2021**, *174*, 105755. [CrossRef]
156. 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]
157. Yu, X.; Li, W.; Gupta, V.; Gao, H.; Tran, D.; Sarwar, S.; Chen, Z. Current Challenges in Efficient Lithium-Ion Batteries' Recycling: A Perspective. *Glob. Chall.* **2022**, *6*, 2200099. [CrossRef]
158. Forti, V.; Baldè, C.P.; Kuehr, R.; Bel, G. *The Global E-Waste Monitor 2020: Quantities, Flows and the Circular Economy Potential*; United Nations University (UNU)/United Nations Institute for Training and Research (UNITAR)—Co-hosted SCYCLE Programme, International Telecommunication Union (ITU) & International Solid Waste Association (ISWA): Bonn, Germany; Geneva, Switzerland; Rotterdam, The Netherland, 2020.
159. Zubi, G.; Dufo-López, R.; Carvalho, M.; Pasaoglu, G. The Lithium-Ion Battery: State of the Art and Future Perspectives. *Renew. Sustain. Energy Rev.* **2018**, *89*, 292–308. [CrossRef]
160. Rallo, H.; Benveniste, G.; Gestoso, I.; Amante, B. Economic Analysis of the Disassembling Activities to the Reuse of Electric Vehicles Li-Ion Batteries. *Resour. Conserv. Recycl.* **2020**, *159*, 104785. [CrossRef]
161. Kamateros, G.; Abdoli, S. Automated Disassembly of Lithium Batteries; Methods, Challenges, and a Roadmap. *Procedia CIRP* **2023**, *119*, 1216–1221. [CrossRef]
162. Wang, S.; Yu, J. Evaluating the Electric Vehicle Popularization Trend in China after 2020 and Its Challenges in the Recycling Industry. *Waste Manag. Res.* **2021**, *39*, 818–827. [CrossRef]
163. Wang, D. Research on Policies of Power Batteries Recycle in China from the Perspective of Life Cycle. *J. Environ. Eng. Landsc. Manag.* **2021**, *29*, 135–149. [CrossRef]
164. Giosuè, C.; Marchese, D.; Cavalletti, M.; Isidori, R.; Conti, M.; Orcioni, S.; Ruello, M.L.; Stipa, P. An Exploratory Study of the Policies and Legislative Perspectives on the End-of-Life of Lithium-Ion Batteries from the Perspective of Producer Obligation. *Sustainability* **2021**, *13*, 1154. [CrossRef]
165. Xu, P.; Tan, D.H.S.; Chen, Z. Emerging Trends in Sustainable Battery Chemistries. *Trends Chem.* **2021**, *3*, 620–630. [CrossRef]
166. Bravo Diaz, L.; He, X.; Hu, Z.; Restuccia, F.; Marinescu, M.; Barreras, J.V.; Patel, Y.; Offer, G.; Rein, G. Review—Meta-Review of Fire Safety of Lithium-Ion Batteries: Industry Challenges and Research Contributions. *J. Electrochem. Soc.* **2020**, *167*, 090559. [CrossRef]
167. Liang, X.; Xue, D. Electronegativity Principles in Metal Oxides Based Supercapacitors. *Nanotechnology* **2019**, *31*, 074001. [CrossRef] [PubMed]
168. Poudel, M.B.; Kim, H.J. Synthesis of High-Performance Nickel Hydroxide Nanosheets/Gadolinium Doped-α-MnO_2 Composite Nanorods as Cathode and Fe_3O_4/GO Nanospheres as Anode for an All-Solid-State Asymmetric Supercapacitor. *J. Energy Chem.* **2021**, *64*, 475–484. [CrossRef]
169. Poudel, M.B.; Kim, H.J. Confinement of Zn-Mg-Al-Layered Double Hydroxide and α-Fe_2O_3 Nanorods on Hollow Porous Carbon Nanofibers: A Free-Standing Electrode for Solid-State Symmetric Supercapacitors. *Chem. Eng. J.* **2022**, *429*, 132345. [CrossRef]
170. Lunde, P. Facilitating Circularity in the Lithium-Ion Battery Value Chain by Designing for Recycling. Master's Thesis, Industrial Design Engineering, Chalmers University of Technology, Gothenburg, Sweden, 2021.

171. Jin, S.; Mu, D.; Lu, Z.; Li, R.; Liu, Z.; Wang, Y.; Tian, S.; Dai, C. A Comprehensive Review on the Recycling of Spent Lithium-Ion Batteries: Urgent Status and Technology Advances. *J. Clean. Prod.* **2022**, *340*, 130535. [CrossRef]
172. Zhang, L.; Zhang, Y.; Xu, Z.; Zhu, P. The Foreseeable Future of Spent Lithium-Ion Batteries: Advanced Upcycling for Toxic Electrolyte, Cathode, and Anode from Environmental and Technological Perspectives. *Environ. Sci. Technol.* **2023**, *57*, 13270–13291. [CrossRef]