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Review

Review analysis of the technology on recycling processes for EV batteries

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ABSTRACT

The increase in use and demand for electric vehicles (EVs) has surged the need for battery recycling methods for these batteries. This report highlights a review analysis of a few recycling methods for EV batteries, such as direct recycling, mechanical recycling, hydrometallurgical recycling, and pyrometallurgical recycling. The purpose of this review is to understand the current state of the technology, the challenges of each method, and the future developments while considering factors such as efficiency, cost, waste production, and more. Direct recycling is reusing EV batteries without disassembling them, whereas mechanical recycling entails discharging, dismantling, crushing, and sorting them. Hydrometallurgical and pyrometallurgical recycling processes both give considerable improvements in metal recovery, with hydrometallurgical recycling including acid leaching and pyrometallurgical recycling using metal extraction. Analyzing the various recycling methods for EV batteries, the effort to improve or innovate the methods will help achieve a more sustainable and effective method to address the EV battery waste, which promotes a circular economy.

1. Introduction

1.1 Background information on EV batteries and their composition

The demand for energy efficiency and environmental awareness is driving up interest in Electric Vehicles (EVs). Carbon dioxide emissions from petrol and diesel-powered vehicles are significant contributors to global warming [1]. Additionally, since EVs are eco-friendly and require clean, renewable energy sources to run, they are a feasible substitute for current fuel-powered vehicles due to the problem of rising global air pollution and diminishing fuel sources [2]. Consequently, research and development of batteries for use in electric and hybrid vehicles are gaining attention [3]. The battery is an essential component because EVs significantly rely on it to store the energy that powers the vehicle [4]. The most popular forms of rechargeable batteries are those made of Nickel-Cadmium (Ni-Cd), Nickel Metal Hydride (Ni-MH), Lead-Acid batteries, and Lithium Ion (Li-Ion) [1]. The most common EV battery components are electrodes (anode and cathode) and electrolytes [5]. EV batteries use lithium transition metal oxide cathode materials such as graphite-$LiMO_2$, $LiMPO_4$, $LiCoO_2$ (LCO), and $LiNiO_2$, with lithium nickel-cobalt-aluminum oxide and lithium nickel-manganese-cobalt oxide batteries being improved versions of $LiMO_2$. Lithium nickel-cobalt-aluminium oxide batteries and lithium nickel-manganese-cobalt oxide batteries are second-generation cathode materials known for their high-temperature thermal performance and minimal capacity loss. There are numerous materials that can be used to create anodes for lithium-ion batteries, including graphite-based (C-based) metal complexes like graphite-$LiMO_2$, Li-TiS_2, Li-MoS_2, and Li-$LixMnO_2$, as well as silicon-based (Si-based) elements found in the Earth's crust [5]. Other anode materials include tin (Sn), cobalt (Co), and molybdenum disulfide. Galvanostatically charging or draining Li-Ion batteries at high currents and low temperatures is made possible by carbon-coated anodes, which increase the capacity of the Li-Ion insertion and extraction. It has been shown that a number of binary solvents, including ethylene carbonate (EC), diethyl carbonate (DEC), ethyl methyl carbonate (EMC), and dimethyl carbonate (DMC), increase conductivity between electrodes and electrolytes in Li-Ion batteries. For Li-ion batteries, lithium salt, also referred to as lithium hexafluorophosphate ($LiPF_6$), is an excellent electrolyte. In order to control the surface chemistry of graphite anodes, advanced active additives such as lithium-bis-oxalato-borate (LiBOB), vinylene carbonate (VC), propargyl-methylsulfone (PMS), hydrofluoric acid and water (HF/H_2O) scavengers, and biphenyl or other aromatic compounds have been added to electrolyte solutions. To enhance the electrochemical and safety performance of solid

Li-ion batteries, novel materials such as gel, polymeric, and glassy matrices have been developed as their electrolytes. Researchers have also suggested high salt-to-solvent ratio electrolytes, solvation-structure ester electrolytes, and composite electrolytes to improve the cycle stability, safety performance, and Coulombic efficiency of Li-ion batteries. Components of a Lithium-Ion battery are shown in Figure 1.

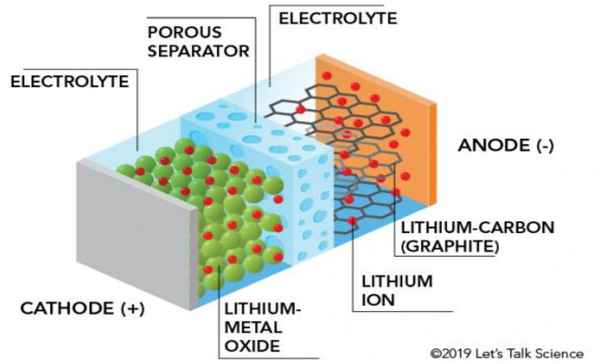

Figure 1. Parts of a lithium-ion battery [6]

1.2 Overview of the importance of recycling EV batteries and their technologies

Due to the global movement to minimize carbon emissions resulting in the rise of the demand for electric vehicles (EVs), the usage of EVs on the road also increases. Hence, the challenge of managing exhausted batteries arises. Batteries for EVs will soon become a major problem if they are not treated properly. This is because extremely dangerous compounds are present, endangering both ecosystems and the people who manage them [7]. Recycling these components is crucial for both environmental and strategic reasons because battery cells' active components also contain important metals, including copper, nickel, lithium, and cobalt. Disposal is doubly expensive because the battery is a substantial cost component for EVs, especially if the waste contains valuable components [8]. By recovering high-value materials and lowering the expense of disposing of hazardous trash, recycling enables the reduction of life cycle costs. Figure 2 illustrates the recycling process of an EV battery. EV batteries must go through multiple processes before they can be recycled because of their complex structure and variety of materials. They must first be categorized and, in most cases, pre-treated via discharge or inactivation, disassembly, and separation before undergoing direct recycling, hydrometallurgy, pyrometallurgy, or a combination of processes, which are some of the available EV battery recycling technologies today [9]. These recycling technologies often include leaching, separation, extraction, and precipitation of electrochemical components [10]. However, EV battery recycling is still in its early phases of development. Hence, much more research and development are required to increase the efficiency, sustainability, and cost-effectiveness of EV battery recycling technologies.

1.3 Purpose and scope of the review

The purpose of reviewing and analyzing technology in EV battery recycling processes is to understand the current state of the technology, identify the challenges of each recycling technology, and estimate the potential for future developments. This type of analysis is critical for informing policymakers, industry stakeholders, and academics on the most effective techniques for EV battery end-of-life management. The scope of this review includes three main recycling technologies for EV batteries, which are pyrometallurgical recycling, hydrometallurgical recycling, and direct recycling.

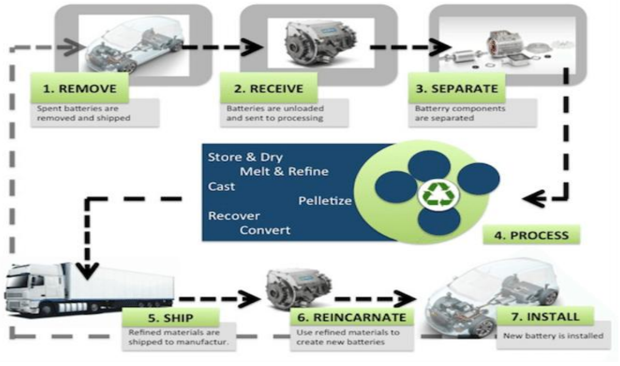

Figure 2. Recycling process of EV battery [11]

2. Mechanical and Pre-treatment process of EV batteries

The mechanical process, also called the physical process, is one of the traditional processes to recycle lithium-ion batteries. This process is a pre-treatment process that is common to all the recycling routes. The mechanical process includes discharging, battery disassembling, and separating the lithium-ion batteries. It concentrates on the valuable bits while separating the battery shell from the other elements. From this process, it is possible to recover materials such as plastics, aluminum, copper, and black matter, which have critical metals, and to be collected for another separate recycling process [12]. Furthermore, the discharge step must be conducted first because the collected battery usually has a specific residual voltage. This can certainly cause spontaneous combustion and explosion if not treated properly, jeopardizing the operators' safety [13].

2.1 Mechanical and Pre-treatment Process

The used Lithium-ion batteries can be physically discharged in the initial step of the process, including forceful discharge and short-circuit discharge. To release the remaining battery power instead, chemical discharge can be performed by submerging the battery in a conductive salt solution. The oxidation-reduction interaction between the positive and negative electrodes is used in this way of discharging to just slightly utilize the remaining battery power. This deactivation step assists in lowering the electrical and flammable risk of recycling the spent batteries; however, it can be neglected if the pyrometallurgy process is continued after the mechanical process. In contrast to the physical discharge method, the chemical discharge method has an advantage due to its high discharge efficiency and quick cycle. This advantage will be useful for a large-scale application. The batteries must then be disassembled and sorted according to the discharge method. The different battery parts can be separated manually or mechanically. The waste battery shell is removed to gather the battery core coil before manually disassembling it. After that, the positive electrode, negative electrode, and organic diaphragm are separated from the battery core coil. For large-scale operation and cheaper cost, mechanical treatment is preferable to hand dismantling, indicating increased economic applicability. After the batteries have been taken apart, they are crushed into positive materials, negative materials, aluminum foil, copper foil, plastic separator, and other pieces [13]. The mechanical procedure for using Lithium-ion batteries is depicted in Figure 3.

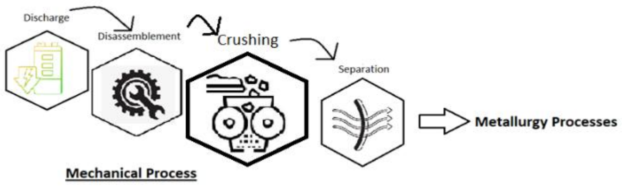

Figure 3. Mechanical process of spent lithium-ion batteries

2.1.1 Case studies of mechanical process

Research on mechanical separation and vacuum metallurgy for recycling the metals from Lithium-ion batteries was done by Zheng et al. [14] in 2018. This study proposed that combining the mechanical recycling process with the vacuum metallurgy as a single integrated process could deal with the bulk amount of spent lithium manganese batteries (18650 $LiMn_2O_4$) without using any additives. Due to residual power in the used batteries, the batteries were thoroughly drained for 24 hours with a five-weight percent NaCl solution before being allowed to naturally air dry. To avoid harming the crusher that would subsequently be utilized in the mechanical recycling process, the hard ion shell was also disassembled, and the integrated process can then start from this point. The pre-treated batteries are then crushed by the crusher before being distributed according to their various particle sizes and being inspected. The spent batteries will next go through mechanical separation for the mixed electrode materials. These materials are thermally treated and are oxygen-free, where they are heated to produce materials that are easy to recycle. The organic binder was eliminated throughout this heating process in the form of a collectible gaseous sample. After 20 to 30 minutes of water leaching, the lithium resource was recovered as lithium carbonate, and the combined electrode materials' valuable metals were recycled after that. The graphite in the filter residue was burned away to recover the manganous-manganic oxide (Mn_3O_4). From the mechanical separation process, crushed pre-treatment batteries were separated by three different diameters. The medium diameter of 0.12 – 0.8 mm was made up of tiny fragments of membrane, cells, aluminum, and copper foil. While cells, membrane strips, copper foil fragments, and aluminum foil strips made up the biggest diameter, which was greater than 0.8 mm.

The largest diameter (>0.8 mm) was composed of copper foil fragments, aluminum foil strips, membrane strips, and cells, whereas the medium diameter (0.12– 0.8mm) was tiny fragments of copper foil, aluminum foil, and membrane. The fine powder (<0.12mm) was mixed electrode materials where this research resulted in obtaining 11.69g of mixed electrode materials from 1 spent 18650 $LiMn_2O_4$ battery, which weighs 36.27 +/- 0.50g with a weight ratio of 32.23%. From the separation obtained, the different diameter particles were analyzed, showing that fine powders were composed of anode powders "graphite" and cathode powders "$LiMn_2O_4$". By recycling 100 spent batteries, the hammer crusher was able to obtain 1185g of mixed electrode materials, showing that the mechanical separation process is an effective process. Table 1 shows the main chemical composition of the mixed electrode materials obtained. The results indicate that the majority of the mixed powers were graphite and $LiMn_2O_4$, while the other trace elements, such as Cu, Al, Fe, Co, and Ni, were included.

Table 1. Chemical composition of mixed electrode materials

Elements	Content (wt.%)
Li	2.371
Mn	37.22
Cu	0.2307
Al	0.2276
Fe	0.0627
Co	0.0095
Ni	0.0062
C	30.83

3. Recycling methods for EV batteries

3.1 Pyrometallurgical recycling

Metallurgy is the science of extracting metals in their pure form for use. Pyro means fire, heat, or high temperatures. Pyrometallurgy is based on heating, extracting, and purification processes used to extract metals from ore [15]. The benefits of pyrometallurgical recycling include its enormous treatment capacity, high chemical reaction rate [16], reasonably flexible feed material, uncomplicated operation, and minimal environmental impact on the slag [14].

3.1.1 Process of Pyrometallurgical Recycling

Pyrometallurgy generally involves these four main processes: discharging, dismantling, pre-treatment, and extractive metallurgy, as illustrated in Figure 4 below. The EV battery assumed here is the standard Lithium-ion battery [15]. Extractive metallurgy can be used to recover enriched metal fractions that are produced as a result of thermal pre-treatment techniques used to break down EV battery modules [17]. It would involve a safe decomposition of combustible and controlled deactivation of organic components of the battery [13]. As the battery's energy content may result in harmful chemical reactions, thermal pre-treatment is also vital for discharging through the disassembly of the battery [18]. Due to the ease with which the cathode materials can be separated via sifting at high temperatures, this approach also eliminates the organic binder materials. Thermal pre-treatment methods consist of incineration and pyrolysis pre-treatment. After pre-treatment, extractive pyrometallurgical methods are deployed to recycle the spent Lithium-ion battery [19]. These methods are roasting/calcination and smelting. Roasting/Calcination is heating compounds in air and transforming sulfide ores into oxides, creating gas [20]. Smelting is used in furnaces for metal reduction and typically involves the formation of carbon dioxide, reducing iron ore in a blast furnace [21]. The last stage is the Refining and purification, whereby leaching, Spray pyrolysis, and Carbothermic reduction (CTR) are used.

3.1.2 Case studies of pyrometallurgical recycling

Glencore Xstrata (Switzerland) recycles spent LIBs as a secondary feedstock by utilizing the pyrometallurgical process [22]. It sees batteries as a specialist market, even though they only make up a small fraction of its overall output [23].

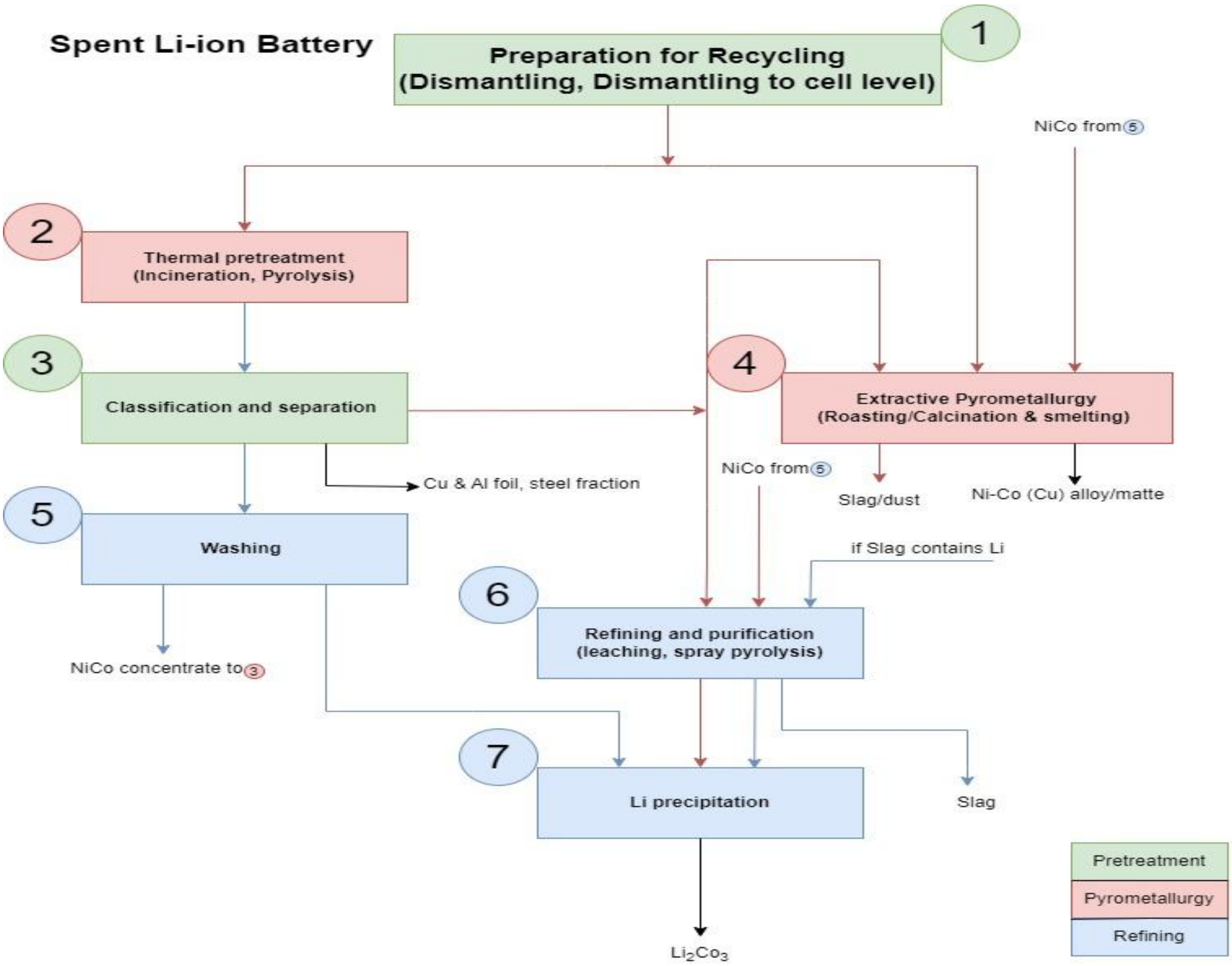

Figure 4. Schematized LIB battery of pyrometallurgical recycling [12]

All LIBs are recycled using hydrometallurgy, pyrometallurgy, and mechanical pretreatment in the recycling process at Accurec Recycling process at Accurec Recycling GmbH in Germany. The battery cells are dispersed, and pyrolysis is used to completely remove all organic components, such as plastics, electrolytes, and binders [24]. The metal components' states are not altered, and the pyrolysis temperature is kept below 250 degrees Celsius [25, 26]. During mechanical pre-treatment, there is no possibility of electrolytes reacting with the atmosphere or fluorine compounds being emitted into the air, which enables Accurec to securely deactivate and destroy combustible organic material [24]. Table 2 shows companies implementing the pyrometallurgical process in LIB recycling. One thing to note is that these are the leading players in the market, but not all worldwide companies are listed here.

3.1.3 Challenges of pyrometallurgical recycling

Due to the high energy consumption and intricate off-gas treatment process, pyrometallurgy requires abundant financing. As research is still being done to develop a recycling system that uses resources efficiently and produces low off-gas, mild hydro-metallurgical (acid-free or alkali) and processing conditions are used in pyrometallurgical recycling as an alternative, which uses intermediate temperatures (<1000^{0}C). Lithium cannot be recovered using the majority of conventional industrial pyrometallurgical techniques [36].

Lithium is a precious mineral because of its dearth and erratic distribution in the Earth's crust [37]. Since other metals like cobalt (Co) and Nickel (Ni) are recovered, recovering Lithium from the electrolyte and lithium metal oxide would be useful [38]. Low recycling efficiency is caused by some minerals that are not recovered. Due to the massive number of used batteries that must be recycled, pre-treatment recycling facilities are eventually incompatible from a technical standpoint [19]. Furthermore, complex changing designs used in EV battery production provide automation challenges and make recycling more difficult.

3.2 Hydrometallurgical recycling

This section discusses the general process of hydrometallurgical recycling, its recycling challenges, and case studies of hydrometallurgical recycling.

3.2.1 Process of hydrometallurgical recycling

In general, hydrometallurgy is a branch of metallurgy that involves the use of aqueous solutions to extract metals from ores or recycled materials. Hydrometallurgical processes have gained significant attention in recent years due to their potential for sustainable metal recovery and environmental benefits. This literature review aims to provide an overview of the existing research on hydrometallurgy recycling processes, focusing on the extraction and recovery of metals from various waste streams. Several studies have explored different leaching agents and conditions to dissolve metals from electronic components. For example, acids such as sulfuric acid, nitric acid, and hydrochloric acid have been widely used [39]. Figure 5 illustrates the simple flowsheet of the hydrometallurgical recycling process of spent LIBs. Researchers have investigated the effects of variables such as temperature, concentration, and leaching time on metal dissolution efficiency [41]. Furthermore, the recovery of specific metals like gold, silver, copper, and palladium has been a subject of interest, and various strategies have been proposed to optimize their extraction [42]. The choice of leaching agent plays a crucial role in the efficiency and selectivity of metal recovery. In addition to traditional acids, alternative lixiviants like organic acids, complexing agents, and deep eutectic solvents (DES) have been explored. Researchers have examined the leaching mechanisms and kinetics of these agents to understand the underlying chemical reactions [43]. The identification of suitable lixiviants and their optimal conditions is essential for maximizing metal recovery while minimizing environmental impact. After metal dissolution, separation and purification steps are necessary to isolate and recover individual metals, as shown in Figure 5. Various techniques have been investigated for this purpose, including solvent extraction, ion exchange, precipitation, and membrane processes. Solvent extraction, in particular, has been extensively studied for its ability to selectively separate metals from complex leach solutions [44]. The optimization of extractants, organic diluents, pH control, and stripping agents has been investigated to enhance the efficiency of solvent extraction processes. Hydrometallurgical recycling processes are often considered more environmentally friendly compared to traditional pyrometallurgical methods. Researchers have focused on minimizing the environmental impact of hydrometallurgy by studying the recycling of lixiviants, reducing reagent consumption, and developing alternative reagents. Additionally, the treatment and disposal of leach residues and effluents generated during the process have been investigated to ensure proper waste management and prevent pollution [45]. Apart from environmental considerations, the economic feasibility of hydrometallurgical recycling processes is a crucial aspect. Researchers have conducted techno-economic analyses to evaluate the overall cost of metal recovery and compare it with traditional methods [46].

3.2.2 Case Studies of Hydrometallurgical Recycling

It is worth mentioning that the most economical, simple, and environmentally friendly hydrometallurgical technique for metal recovery is acid leaching [47]. As a result, case studies on the effectiveness, expense, and energy usage of the hydrometallurgy recycling process for EV batteries are presented in this section. A case study by Chen et al. [22] focused on the hydrometallurgical recovery of metals from lithium-ion batteries used in electric vehicles (EVs).

Table 2. Overview of companies using pyrometallurgical battery-recycling processes [27]

Company	Operational scale	Operating Capacity (t/a LIBs)	Recovered materials (Products)	Recycling processes
Umicore NV (Belgium)	Large scale [28]	7000 [28]	Ni, Co, Cu, Fe, $CoCl_2$ [29]	Pyro, Hydro
Glencore Xstrata (Switzerland)	Small scale (intent to increase Capacity)	7000 [30]	Co, Ni, Cu [24]	Pyro, Hydro [11, 12]
The International Metals Reclamation Company (INMETCO, America)	Commercial scale	6000 [30]	Co, Ni and Fe in iron-based alloy [12, 19]	Pyro, Mechanical [31]
JX Nippon Mining and Metals (Japan)	Commercial scale	5000 [32, 33]	Ni, Co, Li_2Co_3, $MnCO_3$ [34]	Pyro, Hydro [34]
Sony Sumitomo (Japan)	Small scale	150 [35]	CoO	Pyro, Hydro [35]
Accurec Recycling GmBh (Germany)	Medium scale	3000	Li_2Co_3, Co-Alloy	Pyro, Thermal, Mechanical, Hydro
Nickelhütte Aue GmbH (NHA) (Germany)	Large scale	7000	NiCoCu-Matte	Pyro, Thermal, Hydro
Kyoei Seiko (Japan)	Commercial scale	-	Ni, Co, Cu	Pyro
Dowa Holdings Co., Ltd. (Japan)	Large scale	1000	Ni, Co, Cu	Thermal, Pyro, Hydro
SNAM (Societe Nouvelle d'Affinage des Metaux) (France)	-	-	Co, Ni, Cu	Thermal, Pyro, Hydro
Ganzhou Highpower Internation Inc (China)	Large scale	10000	NiMH	Mechanical, Pyro, Hydro

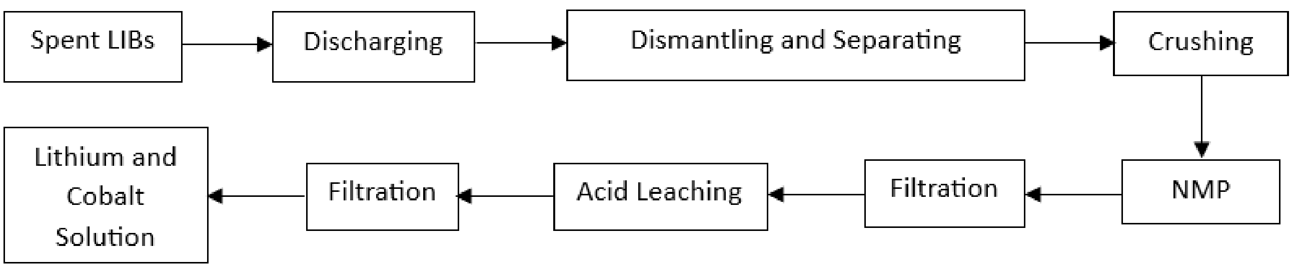

Figure 5. Flowsheet of hydrometallurgical process for spent libs [40]

The study aimed to optimize the leaching process for efficient metal recovery while considering cost and energy consumption. The researchers investigated the use of different leaching agents and conditions to selectively dissolve metals, such as lithium, cobalt, nickel, and manganese, from the battery materials. Their study highlighted the importance of process optimization to achieve high metal recovery rates while minimizing the consumption of reagents and energy. A case study conducted by Choi et al. focused on the recycling of electric vehicle batteries through hydrometallurgical processes [48]. The study aimed to optimize the recycling process to achieve efficient metal recovery while also considering the cost and energy consumption aspects. In order to recover metals like lithium, cobalt, and nickel from depleted EV batteries, the researchers investigated various leaching agents, including sulfuric acid and organic acids. The study emphasized the need for process optimization to enhance efficiency, reduce costs, and minimize energy consumption in the recycling of EV batteries. In a case study by Xu et al. [49], the optimization of the leaching process for cobalt recovery from electric vehicle batteries was investigated. The study focused on enhancing the efficiency of cobalt leaching while considering the cost and energy consumption aspects. The researchers explored different leaching agents, acid concentrations, and process parameters to achieve high cobalt recovery rates. The findings emphasized the importance of process optimization to maximize metal recovery efficiency and minimize reagent consumption and energy requirements. A case study conducted by Sun et al. [50] focused on the energy and environmental assessment of lithium recovery from spent lithium-ion batteries using hydrometallurgical processes. The study aimed to evaluate the efficiency, cost, and energy consumption of the recovery process. The researchers examined different leaching agents and recovery techniques to optimize lithium recovery while considering the energy requirements and environmental impacts associated with each method. The study emphasized the importance of balancing efficiency and sustainability in the recycling of EV batteries. In summary, the case studies contribute to the understanding of efficiency, cost, and energy consumption considerations in hydrometallurgical recycling processes for EV batteries. They highlight the importance of optimizing leaching agents, process conditions, and separation techniques to achieve sustainable and cost-effective recovery of valuable metals while minimizing energy consumption and reducing overall costs.

3.2.3 Challenges of hydrometallurgical recycling

Based on the case studies, several challenges can be identified in hydrometallurgy recycling processes for EV batteries. These challenges include selective metal recovery, process optimization, environmental impact, energy consumption, and cost considerations. One of the key challenges is achieving selective metal recovery from complex battery materials. EV batteries contain a variety of metals, and optimizing the leaching process to selectively dissolve specific metals while avoiding the dissolution of others can be challenging. Hence, developing efficient leaching agents and optimizing process conditions are crucial for achieving high metal recovery rates. Hydrometallurgical processes also require careful optimization to maximize efficiency while minimizing costs and energy consumption. Finding the optimal combination of leaching agents, process parameters (such as temperature and time), and separation techniques can be complex. Therefore, process optimization involves balancing the trade-offs between metal recovery rates, reagent consumption, energy requirements, and overall process economics. Further, recycling processes consider the environmental impact associated with hydrometallurgical methods. The choice of leaching agents and separation techniques can have varying environmental implications. Minimizing the use of hazardous chemicals, reducing waste generation, and implementing proper treatment of process effluents are important considerations for sustainable and environmentally friendly recycling processes. Hydrometallurgical processes can be energy-intensive, particularly during leaching, separation, and purification steps. Reducing energy consumption while maintaining high metal recovery rates is a significant challenge. Process optimization, the use of efficient equipment, and the integration of energy-saving measures are essential to minimize the overall energy requirements of the recycling process. Moreover, the cost of hydrometallurgical recycling processes is a crucial factor for their commercial viability. The selection of cost-effective leaching agents, optimization of process parameters to minimize reagent consumption, and efficient separation techniques are necessary to reduce overall costs. To address the challenges in hydrometallurgy recycling processes for EV batteries, several strategies can be implemented. Continuous research and development efforts are essential to tackle selective metal recovery, process optimization, environmental impact, energy consumption, and cost considerations. This involves exploring new leaching agents, optimizing process parameters, and developing innovative separation techniques. The adoption of sustainable chemistry, such as environmentally friendly leaching agents, can minimize the environmental impact. Implementing energy-efficient measures, utilizing advanced equipment, and recovering and reusing energy can help reduce energy consumption [50].

3.3 Direct recycling

Direct recycling is relatively new in recent years, which has been produced on a lab scale to recycle the active components to reproduce new Lithium-Ion Batteries (LIBs). This will promote a circular economy in producing new LIBs. Direct recycling of LIBs involves separating the excellent purity active components in the cathode and anode from used LIBs and regenerating their electrochemical functionality by different physical, chemical, and mechanical processes. Figure 6 shows a recycling process that involves direct recycling. Direct recycling is a promising technique in conjunction with other recycling methods, which makes use of the lithium nickel manganese cobalt oxide-graphite (NMC-G) battery, the most used type of battery. Typically, when the NMC-G battery is spent, about 20% of functional lithium is lost due to parasitic effects, element isolation, and solid electrolyte interface formation. Without destroying the active elements in the electrodes of LIBs, the direct recycling method may restore and extend the active elements [51].

3.3.1 Process of direct recycling

The process of direct recycling is as follows [52]:

1) Spent batteries will be disassembled into cells and discharged using electrolytes.
2) The cells are then treated using supercritical CO_2 to extract the reusable electrolytes.
3) The remaining electrolytes from the cells are calcined, which produces waste.

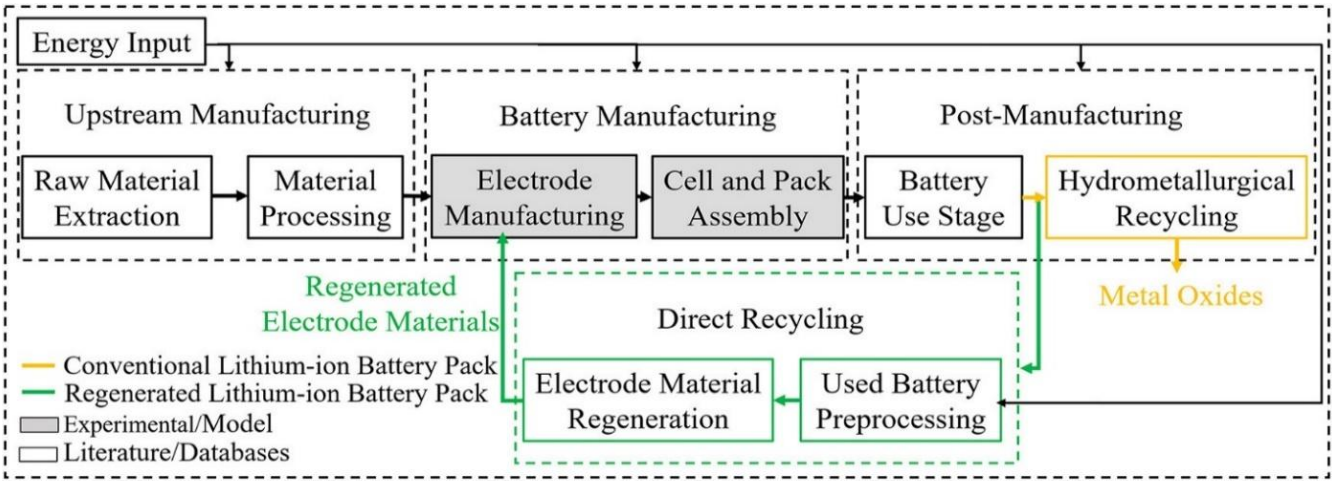

Figure 6. The recycling process includes direct recycling [51]

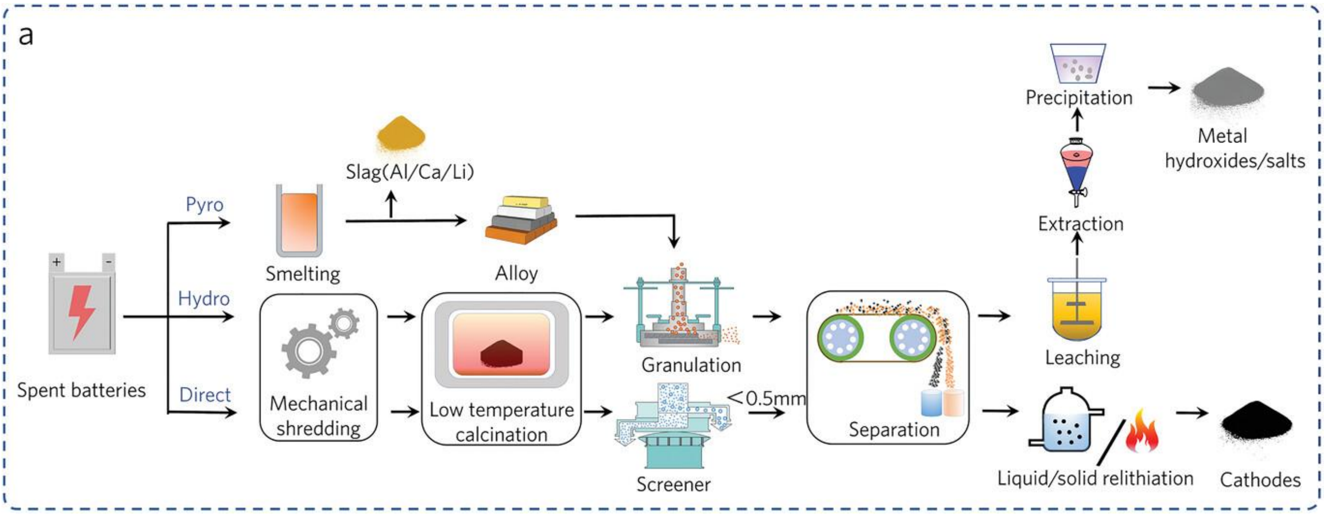

Figure 7. Process of direct recycling [52]

4) The cells are disassembled and pulverized into powder form to separate the materials, such as cathodes, anodes, plastic, and metals, using a non-destructive separation technique.
5) Various re-lithiation methods are used to regenerate the spent materials of the cathodes.
6) Regenerated materials are used in creating new cells for batteries.

Direct recycling is a promising process compared to traditional recycling methods or any existing methods. Figure 7 depicts a process of direct recycling. Compared to pyrometallurgy and hydrometallurgy, direct recycling is comparatively greener and does not have high energy consumption or chemical use. Direct recycling does not cause air pollution or generate much waste, whereby, as mentioned, it focuses on disassembly rather than destruction of the spent battery. Life cycle analysis shows that the regenerated material for the method has a high value in comparison to pyrometallurgy, and hydrometallurgy produces lesser waste emissions and consumes less energy [53]. The NMC-G lithium-ion battery's closed-loop manufacturing methods are depicted in Figure 8. First, presuming that the EVs are similar to regular automobiles at 91%, the used LIB packs are removed from the vehicles.

Around 0.083 MJ of energy is consumed to remove each kg of lithium-ion battery pack. Secondly, the expended LIBs are immersed in a salt solution to remove any remaining charge. Complete discharge of a lithium-ion battery pack is thought to need 0.0035 MJ of electricity per kg. The electrolyte is then extracted from the disassembled battery cell using a CO_2 solvent in the third step, which also involves disassembling the discharged battery cell. The battery cell is fed with compressed liquid CO_2 at a flow rate of 1.5L per minute for about 50 minutes, along with a 3:1 ratio of acetonitrile (ACN) and propylene carbonate mixture at a flow rate of 0.5 mL per minute for about 20 minutes. The carbon dioxide solvent will be converted into gaseous carbon dioxide and extracted from the battery at 100 ml per cell in the electrolyte. Most of the carbon dioxide is reused, whereby the remaining will be considered as recycling consumption. The energy consumption in compressing the carbon dioxide solvent into the cells is roughly 0.04 MJ per kg of the NMC-G lithium-ion battery. Lastly, the cells will be physically reduced and undergo the final process to separate the anode and cathode, which uses 0.26 MJ. The separation process for every battery pack will consume about 0.023 MJ/kg of NMC-G battery pack [51].

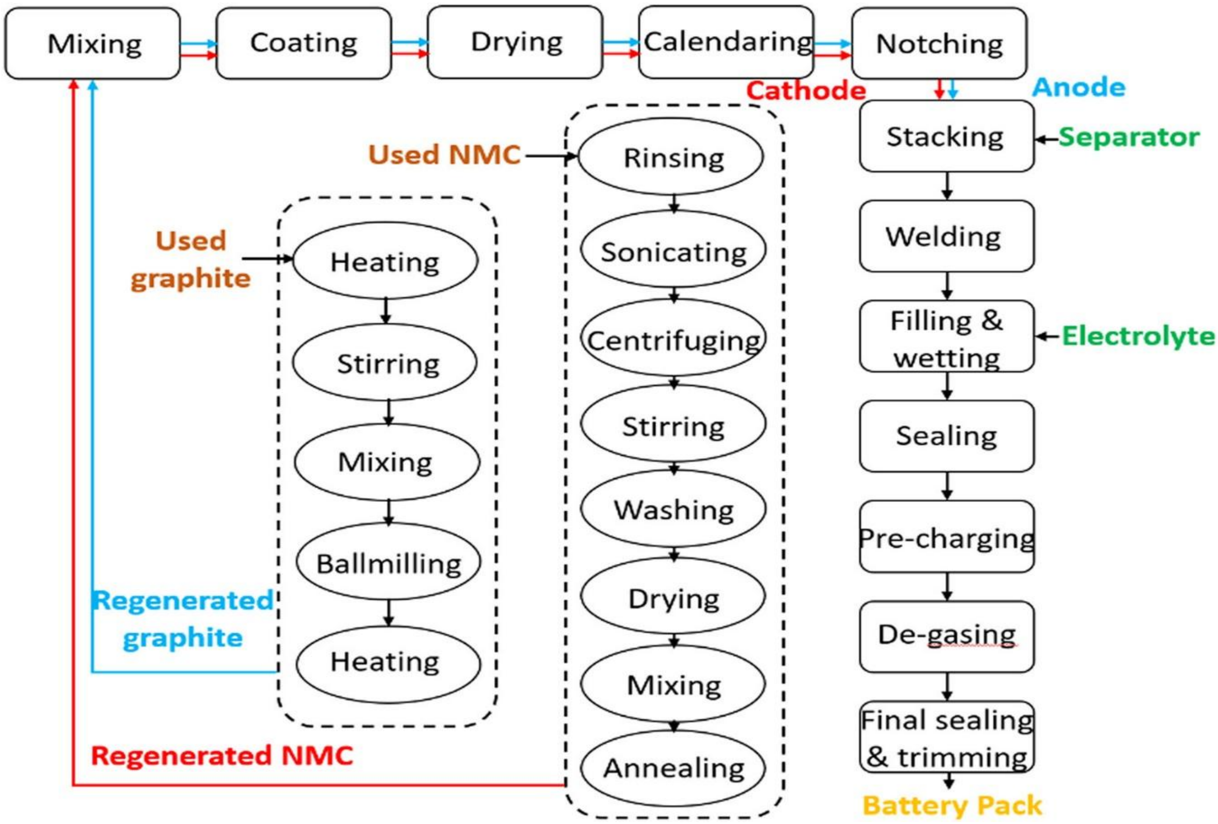

Figure 8. Closed-loop process of recycling spent NMC battery

Direct recycling shows that in countries such as China, South Korea, the US, Belgium, and the UK, it is estimated that direct recycling of various types of EV batteries, such as those from Tesla, proves to have a net profit in comparison to other methods. Figure 9 shows the net recycling profit of various recycling methods. Many factors affect recycling, such as transportation costs, disassembly costs, recycling process costs, the design of the battery, and scale profitability. If direct recycling is able to achieve a similar capacity of recycling compared to hydrometallurgical and pyrometallurgical recycling, then direct recycling will achieve the highest net profit. Direct recycling is predicted to have the lowest break-even point at about 3,000 tonnes per year compared to pyrometallurgical recycling at 17,000 tonnes and hydrometallurgical recycling at 7,000 tonnes per year [54]. On the other hand, Figure 10 shows the potential of direct recycling.

3.3.3 Challenges of direct recycling

Direct recycling is still considered a new process that requires more time and effort before commercialization. The way to improve the method is by addressing the recycling process [55].

1) Preliminary processing in obtaining refined materials: The concept of direct recycling is to obtain the spent materials and directly reuse and regenerate the cathodes. This is prevented as LIBs have many components, such as cathodes, anodes, metals, and plastics. By improving the efficiency of separating the components, direct recycling may be improved, along with ensuring that the retrieved cathodes are of high purity.

2) Recovery of other materials besides cathodes: The direct recycling process currently focuses on the extraction of cathodes in powder form, which is roughly 35% of the cost of the material itself. Without relying on other methods to recover these metals, direct recycling can be maximized even further to reduce the need for other methods.

3) Show the recovered materials: For the EV batteries to take on direct recycling as a better alternative, the method must achieve a certain implementation and establish the importance of direct recycling.

4) Regain the mixture of cathodes: The varying types of spent LIBs may use different cathode substances. This is a challenge for the direct recycling process, as the separation of varying materials is important. Finding a way to extract and separate the various cathode substances may prove difficult, as different LIBs have different ratios of the NMC-G materials. One way is to test whether the mixture of the substances can be extracted directly.

5) Combination of various recycling methods: As direct recycling is still under development, it may be advantageous to implement other recycling methods into direct recycling to allow higher efficiency. Extracted cathode materials may be retrieved using other methods, such as the hydrometallurgical process.

From the previously mentioned discharging process of cathodes using CO_2, the method allows for cathodes to be recycled if allowed. The cathodes may need to be regenerated before being reused in new batteries. This method allows for most components of a spent LIB to be recovered and reprocessed. Cathode materials, regardless of their property or combinations, may be highly valuable through direct recycling. The effectiveness of recovery has yet to be compared to the performance of raw material, which may raise issues later on, such as battery capacity and lifespan. This may spark debate among manufacturers regarding recycled materials, as they have to ensure the product is of quality and performance. Recovered materials may be implemented into other products with less strict requirements [56].

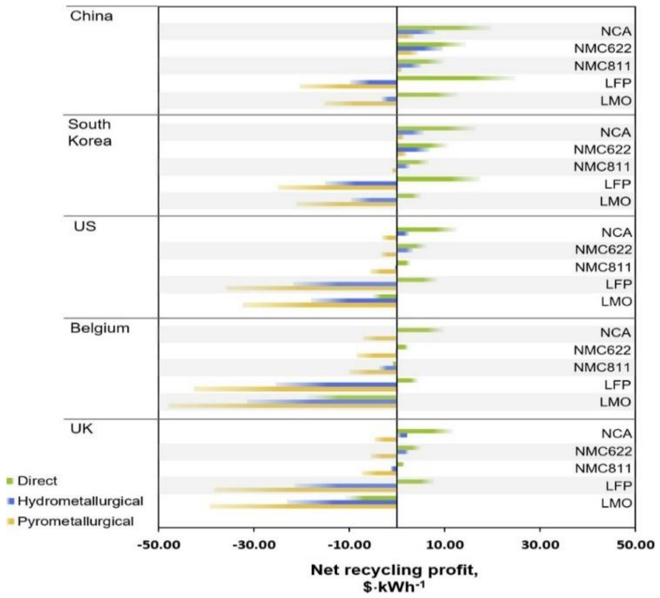

Figure 9. Net recycling profit of various recycling methods [54]

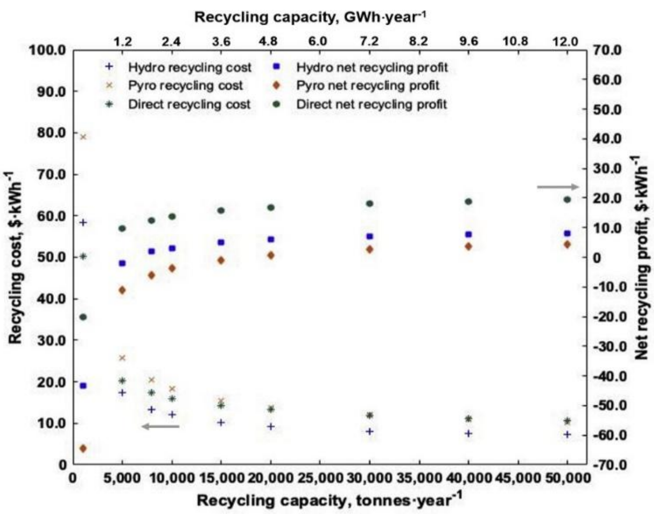

Figure 10. Potential of direct recycling [54]

4. Comparison and discussion of the recycling process

4.1 Comparison table of the recycling process

Table 3 compares the different recycling processes. It compares pyrometallurgical, hydrometallurgical, and direct recycling processes in terms of efficiency, energy consumption, environmental impact, and cost.

4.2 Discussion of the recycling process

Based on Table 3, the efficiency in extracting metals from the waste material of pyrometallurgical recycling is highest compared to hydrometallurgy and direct recycling.

This is because pyrometallurgical recycling involves high-temperature processes such as smelting, where the waste material is melted to separate and recover valuable metals. On the other hand, hydrometallurgical recycling methods use chemical processes to dissolve and extract valuable metals from the waste material. These processes often achieve high efficiency as they can selectively target specific metals and effectively recover them from the solution. Meanwhile, direct recycling involves sorting, cleaning, and reprocessing waste materials to create new products. The efficiency of direct recycling can vary depending on the quality of the waste material and the effectiveness of the sorting and processing techniques. However, with advancements in recycling technologies and improved waste management systems, direct recycling can achieve moderate to high levels of efficiency in recovering valuable materials. Moreover, the energy consumption of direct recycling typically requires low energy consumption. The processes involved, such as sorting, cleaning, and reprocessing, usually require less energy compared to more complex chemical or high-temperature processes. Pyrometallurgical recycling methods have high energy consumption as they involve heating the waste material to high temperatures, which requires significant amounts of energy, whereas hydrometallurgical recycling methods can have moderate to high energy consumption due to the need for chemical reactions and the use of energy-intensive equipment to dissolve and extract metals from the waste material. Besides, direct recycling generally has a low environmental impact compared to pyrometallurgical and hydrometallurgical processes. It avoids the need for extensive chemical processes or high-temperature operations, resulting in fewer emissions and minimal generation of hazardous by-products. Pyrometallurgical and hydrometallurgical recycling methods can have moderate to high environmental impacts. The high-temperature processes involved in smelting generate emissions, including greenhouse gases and air pollutants in pyrometallurgy, and the use of chemicals in the dissolution and extraction processes can generate wastewater and chemical waste, requiring proper treatment and disposal for hydrometallurgy. In terms of overall costs, direct recycling is generally the most cost-effective option. It involves relatively simpler processes and requires less specialized equipment, resulting in lower capital and operational costs. Pyrometallurgical recycling methods have moderate to high costs due to high-temperature equipment, energy requirements, and handling of slag and by-products, which adds to the overall expenses. Hydrometallurgical recycling methods also tend to have moderate to high costs due to the need for specialized facilities, chemicals, and energy-intensive processes. In summary, each recycling process has its own advantages and considerations. Hence, determining which recycling process is better depends on various factors, such as the specific waste material being processed, the desired outcome, and the context in which the recycling is taking place. Direct recycling is generally more cost-effective, has lower energy consumption, and has a lower environmental impact compared to the other methods.

Table 3. Comparison table of pyrometallurgical, hydrometallurgical, and direct recycling processes [57]

Parameters	Efficiency	Energy consumption	Environmental impact	Cost
Pyrometallurgical Recycling	High	High	Moderate to High	High
Hydrometallurgical Recycling	High	Moderate to High	Moderate to High	Moderate
Direct Recycling	Moderate to High	Low	Low	Low

It involves simpler processes and can be more easily integrated into existing waste management systems. However, direct recycling may have limitations in terms of the types of waste materials that can be efficiently processed. It may not be suitable for complex or contaminated materials that require specialized techniques for metal recovery. Meanwhile, pyrometallurgical recycling methods have high efficiency in metal recovery, particularly for metals that can withstand high temperatures. They can handle a wide range of waste materials and can recover a variety of metals simultaneously. Pyrometallurgy is often used for processing large volumes of metal-rich waste. Hydrometallurgical recycling methods, on the other hand, can achieve high efficiency in metal recovery, especially for targeted metals. They can selectively extract specific metals from the waste material, even in low concentrations. Hydrometallurgy is often effective for recovering precious and strategic metals. The limitations of hydrometallurgical methods are that they tend to have higher energy consumption and may generate chemical waste or wastewater that requires proper treatment. They also require specialized facilities and expertise, which can increase costs.

5. Future directions and outlook

The EV battery industry has significant potential for circular economy and sustainability by recycling spent batteries, which can minimize waste and conserve natural resources. To promote EV battery recycling, battery manufacturers should be encouraged to take back waste batteries so that they can be recycled into new batteries or other valuable materials. Policy and regulatory developments should be established to support EV battery recycling efforts, including targets for major components of battery recovery. Recycling initiatives can also include objectives for the utilization of recycled material during the manufacturing of new EV batteries to further promote a circular economy in battery production [58]. EV battery manufacturers globally should be encouraged to implement labeling requirements that provide information on batteries, such as the date of manufacture, chemistry, and hazardous substances, as this can impact recycling initiatives by increasing transparency and facilitating third-party recycling of EV batteries [58]. Moreover, supporting research and development in EV battery recycling technologies would broaden the range of materials that can be recovered and recycled. Through more research and development, recycling technologies have the potential to be more cost-effective and efficient and can be implemented on a larger scale.

6. Conclusion

In conclusion, this paper gives an extensive review of the technologies for recycling electric vehicle (EV) batteries. It emphasizes the significance of efficient battery management due to the presence of hazardous compounds and strategic metals. This is due to the growing EV demand and the need to reduce carbon emissions. The study analyses four major EV battery recycling technologies, namely, mechanical recycling, direct recycling, hydrometallurgical recycling, and pyrometallurgical recycling. Direct recycling involves reusing intact EV batteries or their components without disassembling them, whereas mechanical recycling involves discharging, dismantling, crushing, and separating batteries. Hydrometallurgical and pyrometallurgical recycling techniques provide significant benefits in metal recovery, with hydrometallurgical recycling incorporating acid leaching and pyrometallurgical recycling involving metal extraction using high-temperature processes. Overall, there is an emphasis on the importance of conducting thorough research on the current state of EV battery recycling technologies to make informed decisions about EV battery end-of-life management. Further efforts to conduct research and development of EV battery recycling technologies are necessary to address the challenges of each recycling technique, improve efficiency, and promote the circular economy for EV batteries. The study highlights the significance of sustainable and effective recycling efforts in addressing the environmental issues related to the increasing use of EVs. Valuable materials can be successfully recovered while decreasing costs and reducing the adverse environmental effects of EV battery disposal by promoting the implementation of effective recycling technologies.

Ethical issue

The authors are aware of and comply with best practices in publication ethics, specifically with regard to authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests, and compliance with policies on research ethics. The authors adhere to publication requirements that the submitted work is original and has not been published elsewhere.

Data availability statement

Data sharing is not applicable to this article as no datasets were generated or analyzed during the current study.

Conflict of interest

The authors declare no potential conflict of interest.

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