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A comprehensive review of the reclamation of resources from spent lithium-ion batteries

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ABSTRACT

Due to the increased application of lithium-ion batteries (LIBs), the number of spent LIBs has increased significantly in recent years, which has resulted in new waste management challenges for the recycling industry. The recycling of spent LIBs has gained enormous interest globally, as this can mitigate resource shortages and reduce the detrimental environmental impact of spent LIB waste. As the demand for LIBs continues to grow, it is important to recycle spent LIBs to establish a sustainable supply chain for the critical materials required for battery production. This comprehensive review addresses different strategies for resource recovery from LIBs and covers state-of-the-art processes for recycling LIBs. Additionally, the challenges and strategies for resource recovery from LIBs are highlighted. Furthermore, the advantages and disadvantages of different recycling processes are addressed.

1. Introduction

Rechargeable batteries (also known as secondary batteries) work by passing an electrical current through them in the opposite direction from which they are typically discharged. Unlike primary single-use batteries, rechargeable batteries can be used many times, making them a more economical and environmentally friendly option. Lead–acid, nickel–cadmium (NiCd), nickel–metal hydride (NiMH), and lithium-ion batteries (LIBs) are some examples of rechargeable batteries. Over the past few years, LIBs in particular have drawn a lot of attention. Each battery type has unique characteristics, such as differences in capacity, discharge rate, and lifespan. Rechargeable batteries are employed in various applications, from powering portable electronic devices (e.g., smartphones and laptops) to serving as backup power for uninterruptible power supplies (UPS) in data centers and other critical

infrastructure[1]. Batteries are also used in electric vehicles (EVs) or in hybrid electric vehicles (HEVs) as a source of stored energy to power the vehicle's electric motor.

Notably, while rechargeable batteries offer many advantages over primary batteries, they also have some disadvantages, such as a shorter lifespan and the need to be disposed of properly once they reach the end of their lifetimes. Additionally, improper charging or use can reduce overall performance and lifespan, so it is important to follow the manufacturer's guidelines and best practices when using them. The production of LIBs has increased dramatically in the past few years due to a massive surge in demand for products that use LIBs, particularly EVs, power banks, watches, and mobile phones. Hence, in this review, we focus on LIB-related issues. LIBs are highly acclaimed on a global scale due to their exceptional qualities, such as high specific energy, small size, light weight, good capacity, and low self-discharge rate[2–4]. The

Abbreviations: CAMs, Cathode active materials; CC, Carbon content; Cyt, Cytochrome; DEC, Diethyl carbonate; DMC, Dimethyl carbonate; EBM, Electromembrane; EC, Ethylene carbonate; EMC, Methyl ethyl carbonate; EPS, Extracellular polymers; ERA, Electrochemical recycling approach; EV, Electric vehicle; GHG, Greenhouse gas; HEV, Hybrid electric vehicle; IEA, International energy agency; ILs, Ionic liquids; LCA, Life cycle assessment; LCO, Lithium cobalt oxide; LFP, Lithium iron phosphate; LMO, Lithium manganese oxide; LIB, Lithium-ion battery; LiPF₆, Lithium hexafluorophosphate; LSV, Linear sweep voltammograms; Ni–Cd, Nickel–cadmium; Ni–MH, Nickel–metal hydride; NMC, Lithium nickel manganese cobalt oxide; PG, Purified graphite; PHEV, Plug-in hybrid electric vehicle; PLS, Pregnant leaching solution; PIMED, Polymer inclusion membrane electrodialysis; PVDF, Polyvinylidene fluoride; PTSA, p-toluene sulfonic acid; SG, Spent graphite; SCCs, Spent lithium-ion coin cells; SILs, Supported ionic liquids; TBP, tri-n-butyl phosphate; TCA, Tricarboxylic acid cycle; UPS, Uninterruptible power supplies.

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International Energy Agency(IEA)'s EV30@30 campaign projects that there will be close to 250 million EVs on the road by 2030[5]. The expected annual growth of LIBs for EVs is in the tens to hundreds of billions [5]. According to a 2022 report published by the IEA, the number of EVs on the road reached 16.5 million as a result of sales that nearly doubled to 6.6 million in 2022 from 2020[6]. In 2021, China and Europe collectively contributed to more than 85% of all EV sales, while the US accounted for barely 10% of worldwide EV sales (see Fig. 1(a) and (b)).

2. Composition of LIBs

Anodes, cathodes, electrolytes, separators, and shells are components of all batteries, including LIBs. Various cell types are used, including prismatic, cylindrical, and pouch cells. A current collector (aluminum foil) is covered with active electrode material and a binder to form the cathode. There are a variety of cathode active materials, including Li transition metal oxides (LiCoO_2 , LiNiO_2 , LiMn_2O_4 , $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$, LFP). The crystal structures of the different LIBs' cathode materials are shown in Fig. 2 [7].

The anode consists of an active material and a polymer binder, which are coated in copper foil. For commercial LIBs, graphite is commonly utilized as the anode material, and polyvinylidene fluoride (PVDF) is employed as the binder. A mixture of diethyl carbonate (DEC), ethylene carbonate (EC), dimethyl carbonate (DMC), and/or ethyl methyl carbonate (EMC) serves as the organic solvent for electrolytes, which are composed of electrolyte salts, such as lithium hexafluorophosphate (LiPF_6) (Fig. 3). The shell usually consists of plastic, stainless steel, or aluminum, while the separator is made of a polymer material[8].

Rechargeable battery technologies have advanced because of increasing requirements for sustainable power. After their commercialization, LIBs have supplanted other energy conversion and storage devices. With the improvement of the battery industry, waste batteries have become both a significant secondary resource and a serious cause of pollution[9]. Various materials, including graphite, Li, Co, Al, Mg, Cu, Mn, and Ni, are used in the fabrication of LIBs[4,10]. It is anticipated that supply risks for Li, Co, and graphite could lead to shortages and price volatility[4]. It is estimated that the LIBs sector uses 25% of the Co and 35% of the Li produced globally[11,12]. Given the considerable demand for graphite in the EV industry, the supply of graphite will be further constrained[13].

Furthermore, given that the LIBs industry will continue to expand, concerns have been raised about supply and value chains. The usual lifespan of an LIBs is estimated to be 8 to 10 years, and as a result of their widespread use in EVs, massive quantities of spent LIBs have appeared all over the world[14,15]. If waste LIBs are not properly recycled at the end of their lives, this will result in millions of tons of spent LIB waste, which squanders resources and harms the environment[3,10,14]. It is anticipated that by 2030, approximately 11 million tons of spent LIBs will be generated, which indicates that there is a huge market for the recycling of LIBs[10]. Spent LIBs should be considered a significant secondary resource that can be recycled to mitigate supply risks and reduce the associated harmful environmental impacts. The recycling rate of LIBs varies by region and country, but it is generally estimated to be low. Fig. 4 displays papers pertaining to recycling LIBs waste, demonstrating the growing interest in battery recycling research.

Recycling has significant economic benefits in addition to resource supply risk mitigation advantages[3]. The development of spent LIBs recycling technology has steadily gained attention as the production of EVs has increased. However, even though the volume of spent LIBs is increasing quickly, the recycling of used LIBs is not advancing at the same rate[16]. The global LIBs recycling market size is predicted to reach 22.8 billion by 2030[17]. According to the US Department of Energy, only about 5% of LIBs in the United States are recycled, while the European Union's Battery Regulation aims to boost the recycling rate of batteries to 45% by 2025. Numerous factors, such as the lack of standardized recycling procedures, the relatively high cost of recycling, and the difficulty associated with collecting and transporting spent batteries for recycling, hinder the recycling of LIBs. Additionally, many people are not aware of the importance of recycling batteries or may not know how to properly dispose of used batteries. Recycling facilitates resource conservation, mitigates the environmental impact of battery production, and ensures the safe handling of hazardous materials. Several initiatives are underway to increase the recycling rate of LIBs, including government-led programs and private sector efforts. For instance, the EU's Battery Regulation mandates that battery manufacturers finance the collection and recycling of spent batteries as well as provide information to consumers on how to properly dispose of used batteries[18]. Additionally, a growing number of companies are specializing in the field of LIBs recycling, and many are working to develop efficient and cost-effective recycling processes. Due to the rising

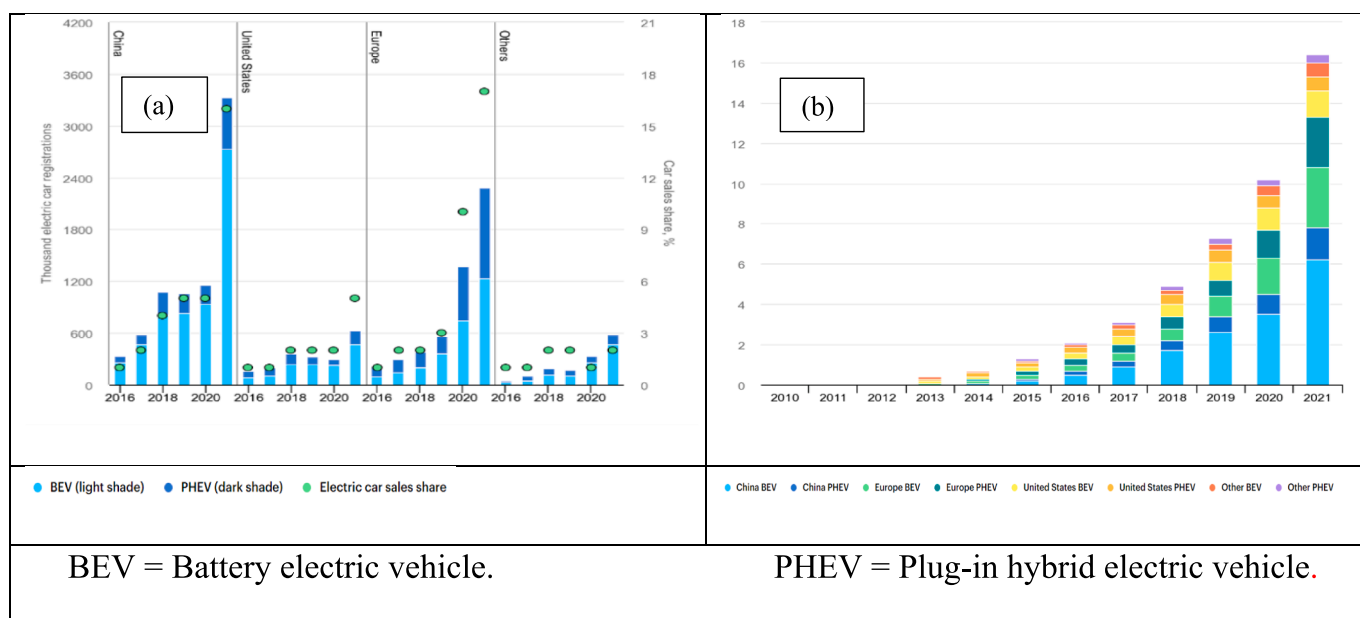


Fig. 1. (a) Registration and sales share of electric cars in China, Europe, US, and other regions, 2016–2021[6] (License: CC BY 4.0); (b) Global electric car stock, 2010–2021[6] (License: CC BY 4.0).

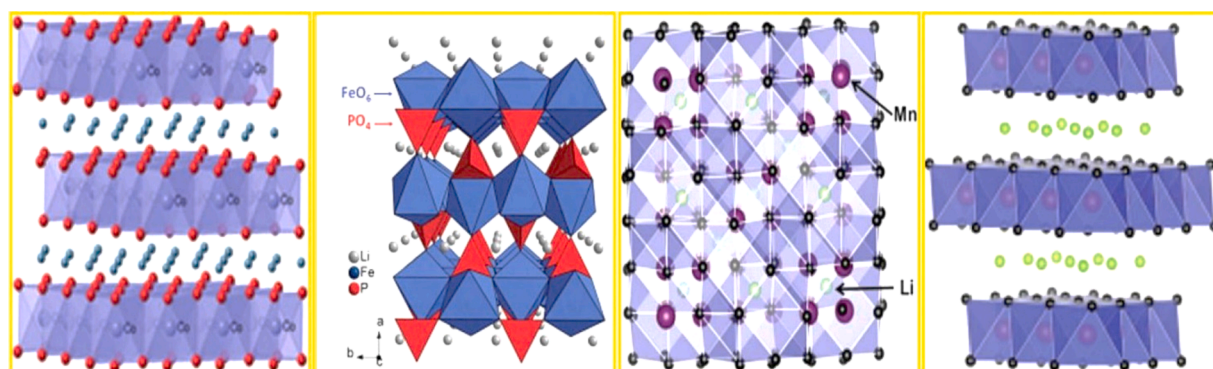


Fig. 2. Crystal structure of different LIB cathodes (a) LCO-Layered structure; (b) LFP-Olivine structure; (c) LMO-Spinel structure (d) NMC-Layered structure [7]. Reproduced with permission from Ref.[7]; Copyright © 2021 American Chemical Society.

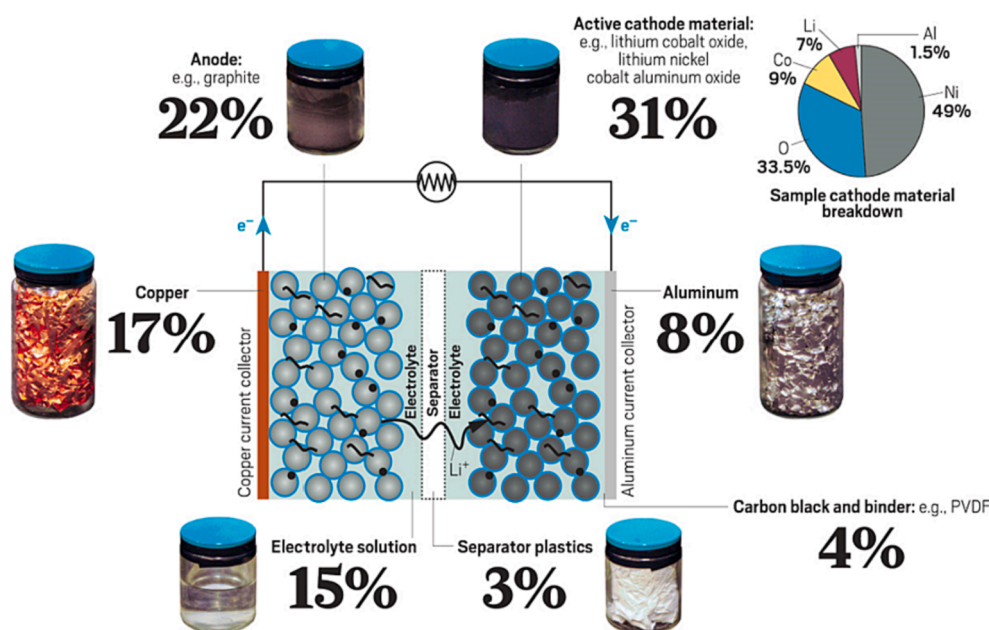


Fig. 3. Composition of LIB cell. Reproduced with permission from Ref. [8].

demand for LIBs and the steadily increasing costs of their constituent materials, including Co, Ni, Li, and graphite, it is crucial to develop efficient approaches for the recycling of LIBs.

3. Environmental aspects of LIBs waste

Landfilling and incineration are the most common approaches in the disposal of spent LIBs[14,19,20]. The incineration process results in the emission of hazardous or even toxic gases, which can have a negative impact on the environment[19,21]. Hydrogen fluoride (HF) is the most dangerous of the toxic chemicals that LIBs discharge into the atmosphere. Battery fires in landfills or in the case of accidents may also emit harmful gases[22]. LIBs' chemical contents can also leach out into the environment[23]. Continued dumping of spent LIBs at landfills could lead to severe environmental issues, such as contaminated water and soil [23,24].

LIBs leachates may contain dissolved gases, heavy metals, additives, electrolyte degradation products, and other types of contaminants. Heavy metals could wash away in the rain and end up in the soil, a neighboring river, or a lake[25,26]. Leachate from landfills is a concern because it can act as a gateway for the dispersion of contaminants beyond landfill sites[27]. In LIBs, cathode materials are typically composed of organic electrolytes that are detrimental to the

environment, and LIBs may leach leftover electrolytes and their additives[14,28]. LIBs may potentially leach electrolyte residues and their additions. Additionally, the electrolytes of spent LIBs comprise hazardous lithium salts and volatile organic compounds, making them susceptible to a variety of chemical reactions when they come into contact with air and water. This poses a major risk to human health and can cause secondary pollution. In terms of both environmental sustainability and the economy, resource recovery from LIBs waste is necessary [8,20,24,29-32].

4. Consequences of recycling spent LIBs

Research and development efforts in the field of sustainability are currently focused on green energy, indicating a shift toward changing energy systems. The valuable materials contained in a product are still present when it reaches the end of its life cycle. These materials can be reused through recycling, thus starting a new technological cycle[33]. Therefore, it is more lucrative to extract metals from waste LIBs than to acquire and utilize pure and genuine metals, since waste LIBs are more cost-effective to acquire, have higher circulation rates, and carry no supply risks. Furthermore, recycling processes can improve waste management and decrease the reliance on imported critical raw materials [34,35]. Reuse, repair, and recovery are the three main ways to

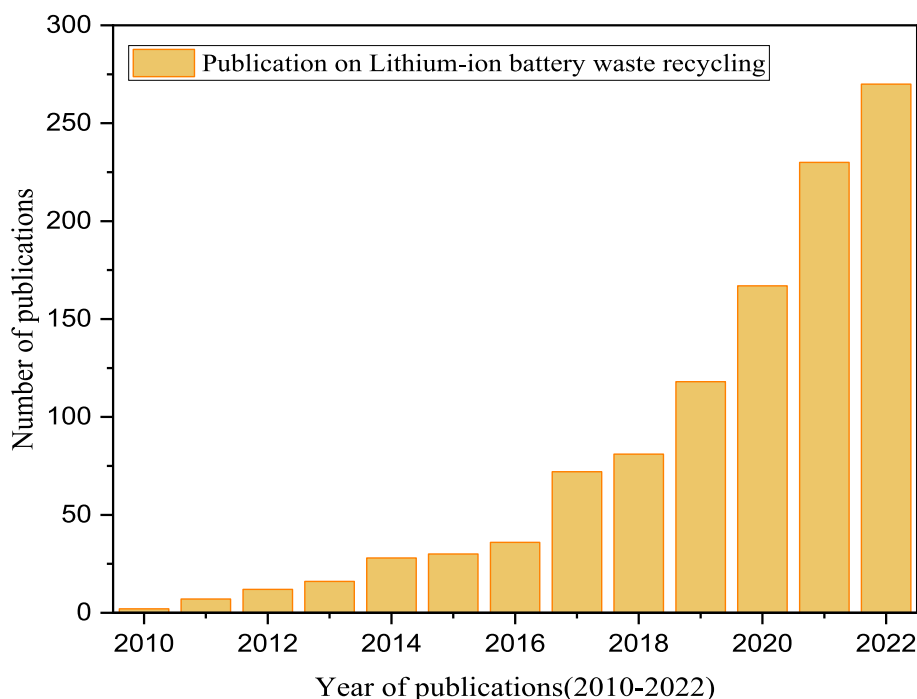


Fig. 4. Web of Sciences: Keywords- Lithium-ion battery waste recycling: (2010–2023): Date of access 28.4.2023.

recycle used LIBs, with recovery being the process that has gained the most attention[20].

From a recycling standpoint, LIBs have many advantages:

- (i) **Resource conservation:** LIBs recycling conserves valuable resources, such as Li, Co, and Ni, by recovering and reusing these materials in the production of new batteries. This reduces the demand for additional resources and the environmental damage caused by resource extraction.
- (ii) **Reduced waste generation:** LIBs' recycling helps to reduce the amount of waste generated at the end of LIBs' life cycles. This is important for reducing the environmental consequences of waste disposal and minimizing the emission of hazardous chemicals and heavy metals.
- (iii) **Energy savings:** The recycling of LIBs can be more energy efficient than the production of new batteries, as it typically requires less energy to extract and refine recycled materials. However, this is strongly case-specific[36]. This helps to minimize greenhouse gas emissions and mitigate the environmental impact of battery production.

5. Spent LIBs waste as a resource factory

The main components of LIBs are electrode materials (cathode and anode), current collectors (Cu and Al), separator, electrolyte solvent, salt, and other materials used in paste formulation and cell assembling, such as conductive agents, binders, and solvents[37]. In the 2030 s, the limited availability of certain elements, particularly Li, Ni and Co in certain regions, will make crucial metal ions, such as Ni, Co, Mn highly valuable[38]. Thus, recycling and recovering valuable elements and metals from LIBs are crucial for many reasons[39,40]. The recycling of spent LIBs primarily depends on hydrometallurgical and pyrometallurgical processes[41] and/or combinations of them.

(a) Cobalt (Co).

Co is a highly valuable metal that is used in various industrial, commercial, and military applications. It is mainly used to produce turbine parts for aircraft engines using superalloys. Magnets, anti-corrosion alloys, diamond tools, catalysts, enamel, pigments, battery

electrodes, and magnetic recording are other applications involving the use of Co[42]. It is one of the key elements of LIBs with the highest economic value[43,44]. The recovery and recycling of Li and Co from used LIBs has been the subject of numerous investigations[45–47]. Even if the relative amount of Co in LIBs is decreasing, the effective recovery of Co from used cathode materials is a significant issue[33]. Nevertheless, it can be challenging to obtain recovered material with integrity similar to the commercial scale. Consequently, the reuse of Co and lithium cobalt oxide (LCO) from LIBs has raised new technological issues [33].

Co is extracted from LIBs with the help of an extractant called Cyphos IL 102, which contains phosphonium ions. With an increase in the Cyphos IL 102 concentration, the proportion of Co extracted increases sharply, and 96% of the Co can be extracted. Therefore, it is economical to use Cyphos IL 102 as an extractant for recovering Co. Cyphos IL-101 has been suggested for the extraction and recovery of used LiCoO₂ battery leach liquors utilizing continuous circulating extraction. Thermodynamic analysis has shown that Cyphos IL-101 has a high capacity and stability for the efficient extraction of Co ions, and a temperature increase facilitated better Co and Li extraction[48].

Another study attempted to recover Co from spent LIBs by utilizing solvent extraction with Cyanex 272 for a real chloride-based solution after Mn removal. To maximize the separation of Co and Ni, the extraction efficiency as affected by various variables was examined. Furthermore, to identify the number of extraction phases required, a new technique utilizing a dynamic approach based on the experimental design was created[49]. In a recent study, valuable Li and Co elements were recovered from LIBs using organic p-Toluenesulfonic acid (PTSA). These elements can be used to create battery raw materials, such as Li₂CO₃ and Co₃O₄. The study demonstrated that using PTSA for leaching could ensure the high recovery of Co₃O₄ and Li₂CO₃ via hydrometallurgical recycling of spent LIBs for practical purposes. The recovery process is simple, cost-effective, environmentally friendly, and crucial for properly recycling spent LIBs.

A study showed that glycine can efficiently extract a high concentration (89%) of Co from cathode materials in LIBs. Optimal leaching conditions studied included the liquid-to-solid ratio and temperature. Co could be recovered from glycine leachates using oxalic acid to create

Co-oxalate residue, which can then be used to make new LIBs[50]. Co recovery from LIBs was examined using various established hydrometallurgy techniques, such as acid leaching, solvent extraction, ion exchange resins, and precipitation. Even though inorganic acid leaching is a commonly used method, the leaching potential of organic acids can be achieved, and the benefits of solvent extraction for achieving higher separation degrees may be less risky, more affordable, and also more sustainable[51].

(b) Lithium (Li).

Ores are the primary source of Li supply, but they are gradually running out due to exploitation[52]. Li is a significant factor in EVs due to its high heat capacity, potential redox value, and impact on modern life[53]. Therefore, there will likely be a significant increase in Li demand, which could reach six times the capacity of current mineral sources, in the next 20 years[52].

Hence, Li extraction from LIBs recycling is beneficial in various ways, such as saving resources, advancing environmental policies, and protecting future generations[30]. A study suggested an improved method for recovering lithium from $\text{Li}(\text{Ni}_x\text{Mn}_y\text{Co}_{1-x-y})\text{O}_2$ cathode materials using carbothermal reduction and water leaching[54]. The carbothermal reduction could recover up to 93% of Li from end-of-life LIBs, with water leaching with a low liquid–solid ratio and graphite in situ, thus reducing energy consumption and costs. Furthermore, using a separator as a carbon source in the recycling process increased Li recovery from 30% to 62%, resulting in a maximum Li recovery of 62% and a Li purity of 92%. Aluminum extracted increased with temperature and time but decreased beyond 650 °C[55]. Despite this, there is still an urgent need for direct conversion solutions to recover lithium from slag (obtained from the pyrometallurgical treatment of LIBs) or spent battery black mass (crushed and shredded LIBs).

(c) Nickel (Ni).

The requirement for Ni in batteries has increased with the rapid evolution of EVs and the tendency to use high-nickel cathode materials. The battery industry consumes the second-highest quantity of Ni after the steel industry, from 7% in 2021 to 37% in 2030[56]. Due to the significant expansion of the battery industry and its potential environmental harm, it is crucial to recycle used nickel[57]. Currently, Ni is recovered via pyrometallurgical and hydrometallurgical processes. The key challenge is the need for pure Ni in the battery industry.

(d) Manganese (Mn).

In addition to Co and Ni, Mn is increasingly being used in batteries. Minerals, manganese nodules, and Lithium manganese oxide (LMO) cathode materials are known sources of manganese[58]. In recent years, LIBs have become one of the most important secondary resources of manganese. The spent lithium-ion coin cells (SCCs), or LMO, are a valuable source of metals[59]. LiMn_2O_4 is significantly cheaper and has excellent safety features. Consequently, LiMn_2O_4 cathode materials are growing in popularity and occupy a larger share of the LIBs market[60]. Furthermore, new manganese-rich compounds, such as LNMO, have been developed and commercialized[61]. Manganese from the metal refining industry can be recovered and purified (e.g., from anode sludge for battery use)[62,63].

(e) Copper (Cu).

Copper is a key element in electronics and is used as a current collector in LIBs. The efficiency of LIX984 extractant was examined to create a novel process to extract copper from a high pH ammonia leaching solution of spent LIBs and regenerate the $\text{LiNi}_{0.5}\text{Co}_{0.5}\text{O}_2$ cathode material[64]. The extracted Cu was stripped using a 1.5 M H_2SO_4 solution. Given the material's excellent electrochemical performance, this process can be an alternate technology for recycling spent LIBs.

(f) Aluminum(Al).

Aluminum is one of the most valuable elements used in LIBs. Aluminum foil is used as a cathode current collector material in LIBs. Furthermore, aluminum may be present in cathode active materials (CAM) due to doping or coating (high-temperature solid-state treatment) procedures. Metallic aluminum foil can be recovered by

separating the foil from the active cathode material. However, due to the strong adhesive effect of the binder, the separation process may be challenging. The separation of cathode active material from aluminum foil can be promoted using suitable organic solvents and ultrasonic-assisted treatments. Aluminum can be recovered effectively by applying alkaline dissolution. Nan et al. showed that 98% of aluminum was dissolved when treated with a 10 % NaOH(w/v) solution at ambient temperature for 5 h.

6. Resource recovery approach

Recycling spent LIBs involves the reclamation of valuable resources from spent batteries and their repurposing in the manufacturing of new batteries. Recycling spent LIBs can be more expensive than conventional waste disposal techniques and requires specialized facilities, processes, and equipment. However, as the demand for LIBs continues to grow, recycling will become increasingly important to conserve valuable resources, reduce the environmental impact of resource extraction, and support the growth of a circular economy for batteries. The recycling process typically involves several steps, as shown in Fig. 5. Resource recovery methods and applications are significantly influenced by the chemistry of the battery materials and the valuables to be recovered. Different approaches used in the recovery of resources, such as metals, electrolytes, and graphite materials, are discussed in this review.

6.1. Electrolyte recovery and collection

Wide attention has been given to the recycling of valuable metals, but electrolyte collection has been neglected. In LIBs, electrolytes contain dangerous lithium salts that can trigger chemical reactions when they come into contact with water or the air, causing secondary pollution and endangering human health. Consequently, it is advantageous and necessary to collect and recycle electrolytes. Supercritical CO_2 extraction and solvent extraction are the two main techniques used to recover electrolytes. The first is used in the preliminary pretreatment stage but is highly inefficient. Supercritical CO_2 extraction results in a more advantageous outcome in collecting electrolytes from battery cells; the supercritical CO_2 fluid acts as an extractant to separate the electrolyte from the electrodes (as highlighted in Fig. 6a). This technique can efficiently extract organic components so that the CO_2 can be further reused. First introduced by Sloop and Allen (2016), this technique can be regarded as a well-established process that successfully reduces hazardous gas emissions[65].

Another work reported using supercritical CO_2 for electrolyte extraction[66]. The recycled electrolyte was compared to a commercial electrolyte, and it displayed high ionic conductivity ($0.19 \text{ mS}\cdot\text{cm}^{-1}$ at 20 °C). Fig. 6b shows the Linear sweep voltammograms (LSV) results for both recycled and commercial electrolytes. Both electrolytes were demonstrated to have stability windows up to 5.4 V. The authors reported that the electrochemical stability of the recycled electrolyte was less than that of commercial electrolytes, but even so, it was sufficient for the operation of electrodes based on LiNiO_2 , LiCoO_2 , and LiMn_2O_4 . Fig. 6c shows the charge/discharge profiles of recycled and commercial electrolytes at a current density of 0.2C. The curves show similar trends, which suggests that the recovered electrolyte exhibited a very good performance similar to that of the commercial electrolyte. The results indicate that supercritical CO_2 is an efficient method for recovering electrolytes with satisfactory electrochemical performance in relation to commercial electrolytes. Recently, another method for recovering electrolyte components was studied. Electrolyte solvents could be recovered using thermal treatment below 150 °C. DMC, EMC, and EC from the exhaust gas were successfully trapped in a mixture of dry ice and acetone at −78 °C. Unfortunately, LiPF_6 could not be recovered. LiPF_6 is not thermally stable and therefore decomposed to $\text{POF}_3(\text{g})$ and $\text{HF}(\text{g})$, which were collected in the water phase and eventually formed $\text{HF}(\text{aq})$ and $\text{H}_3\text{PO}_4(\text{aq})$ [67].

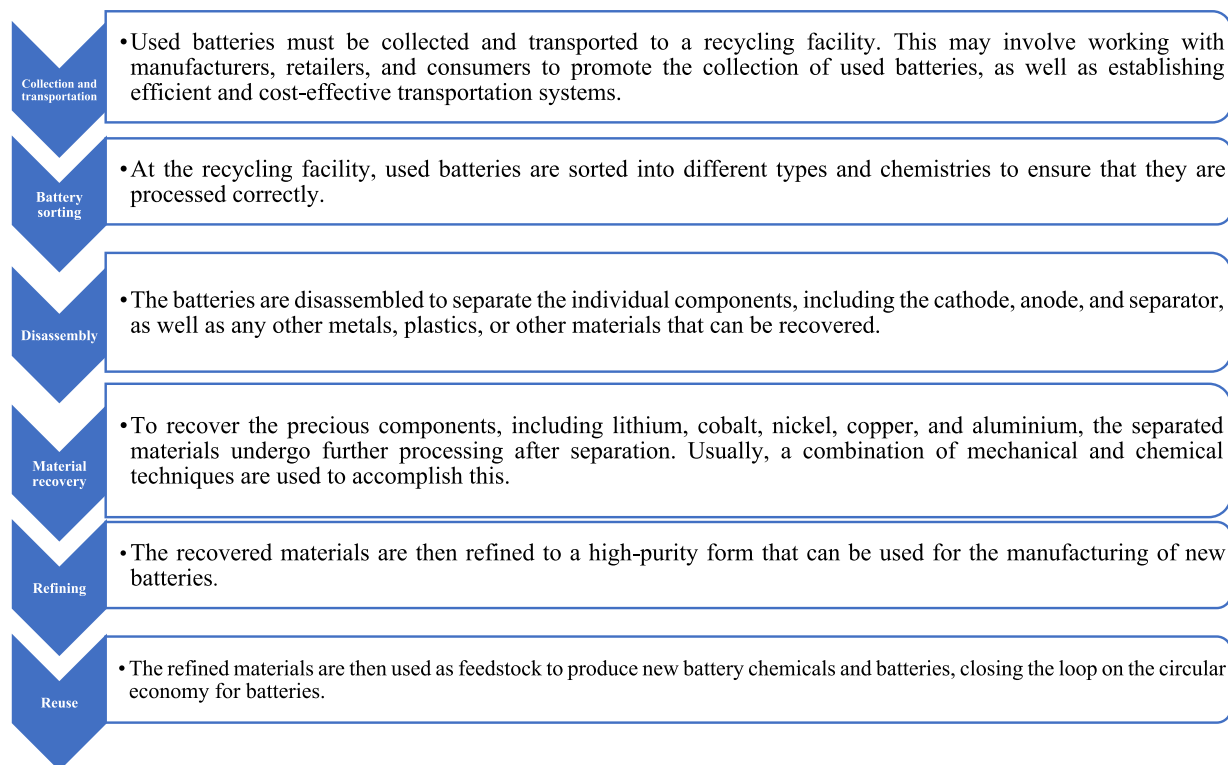


Fig. 5. Different steps in the battery recycling approach.

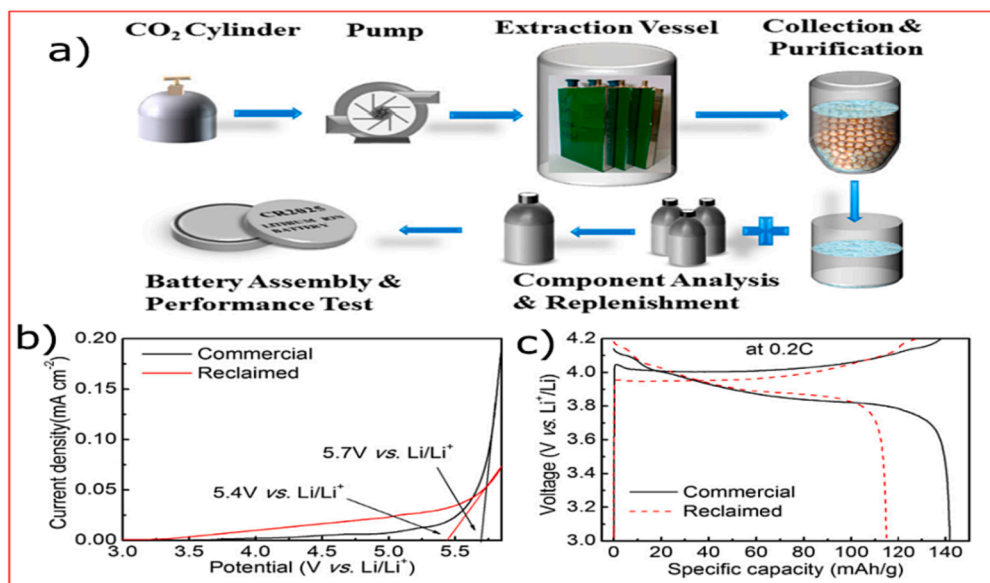


Fig. 6. (a) Schematic of electrolytes recovery from spent LIBs; (b) Linear sweep voltammograms for commercial and reclaimed electrolyte at 1 mV·s⁻¹. (c) Charge/discharge curves of a Li/LiCoO₂ cell at 0.2C with commercial and reclaimed electrolytes. Reprinted with permission from Ref.[66]. Copyright (2017) American Chemical Society.

6.2. Graphite recovery

Graphite is one of the most frequently used anode materials in LIBs. Natural graphite has been classified as a critical raw material by the EU. In the EU, LIBs recycling processes focus on the recovery of Ni, Cu, and Co, while graphite is not currently being recovered[18]. One option to increase the graphite supply in the EU could be recycling and reusing graphite from spent LIBs. The most common techniques for recycling graphite are hydrometallurgical treatments, pyrometallurgical

treatments, or a combination of both. Purification usually starts with heat treatments to remove organics to improve the liberation of electrode materials[68]. Cathode materials are usually removed by acid leaching using acids such as sulfuric acid[69-75] and hydrogen chloride [76]. Hydrogen peroxide has also been used to improve leaching [69,73,76]. Leaching with water[77,78], citric acid[79], and ammonium persulfate[80] has also been studied. High-temperature treatments are used to remove impurities but also to repair the damaged structure of spent graphite (Fig. 7)[70,71]. Others have also utilized microwave

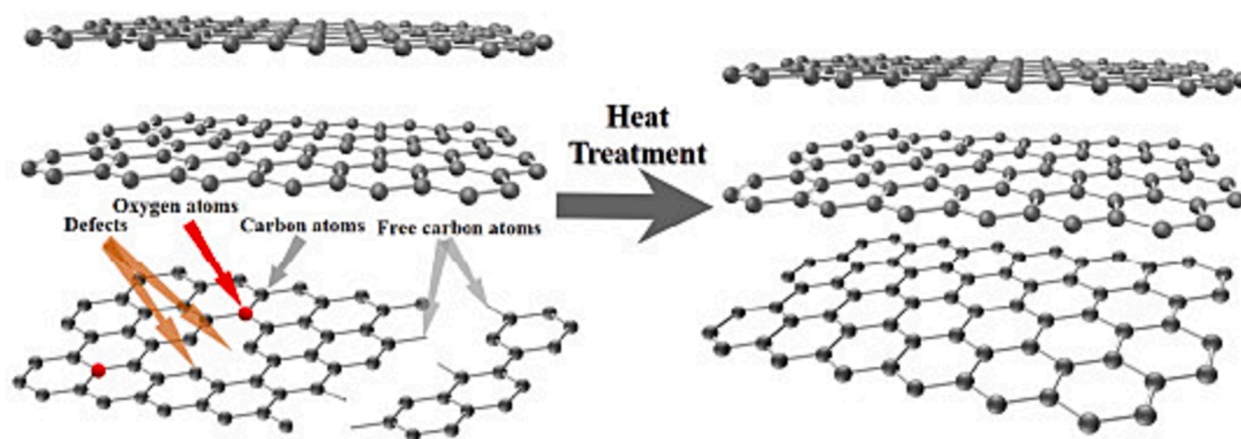


Fig. 7. Regeneration of spent graphite from spent LIB by high temperature treatment. Reprinted with permission from Ref.[71]; Copyright © 2021 Elsevier.

[72,74,81] and carbon coating processes[75,82,83] to regenerate spent graphite. Table 1 presents different purification processes and conditions in which spent graphite has been recycled from LIBs.

Graphite is an essential part of battery anodes, and its production is energy intensive. Traditionally, amorphous carbon is synthesized to graphite using the Acheson thermal process (requiring temperatures of 3000 °C)[84]. Biomass-based carbon has also been studied as an alternative carbon source for graphite production. Biomass-derived materials have several advantages because they are inexpensive, abundant, and have less environmental pollution concerns[84,85].

6.3. Metal recovery

6.3.1. Acid leaching by inorganic and organic acids

Different inorganic acids, such as sulfuric acid[86,87], nitric acid [88], or hydrochloric acid[21], can be used in metal leaching from spent LIBs. Some auxiliary chemicals, such as H_2O_2 , can also be used to improve leaching[86]. In addition to conventional inorganic acids, more environmentally friendly organic acids, such as oxalic, citric, malic, ascorbic, succinic, tartaric, formic, and lactic acid, are used in the

leaching process. As shown in Fig. 8, in the leaching approach, the particles usually undergo a loosening-breaking-shrinking change process[89]. Acid leaching methods to recover different valuables from LIBs using inorganic and organic acids are listed in Table 2 [89–108].

6.3.2. Precipitation

Precipitation can be used to uptake valuable materials in waste LIBs [93]. Simultaneous recovery of Li, Co, Ni, and Mn is quite challenging. Common impurities with relatively high concentrations in the leaching solutions of black mass are Al, Cu, and Fe. A common precipitation method for Al(III) and Fe(III) is pH adjustment to 3.5. Aluminum and iron precipitates as hydroxides have very low solubility at pH 3.5.[35]. Sulfide precipitation is a selective method used to recover CuS precipitate. CuS has low solubility, even under very acidic conditions. Acid + H_2O_2 based leaching of battery black mass can generate sodium sulfite in the precipitation stage. Kang et al. used Na_2S to remove copper from LIBs leaching solutions and to do this, they added 3:1 S:Cu at pH 1 and 25 °C. A 99.9% removal of copper was achieved[110]. Furthermore, manganese could be recovered by oxidative precipitation using $KMnO_4$ [93] or ozone[111] to oxidize Mn(II) to Mn(IV). Schaeffer et al.

Table 1
Recovery of spent graphite from used.

CC _{SG} (wt-%)	Purification process	L/S ratio	t (min)	T (°C)	Structural reconstruction	CC _{PG} (wt-%)	Electrochemical performance of PG	Reference
88.5	1. SG mixed with concentrated H_2SO_4 (1:1) + di-water (10% of total mass), 2. Roasted, 3. Leached with H_2SO_4 (200 g/l)	–	1. 10 2. 120 3. 240	2. 200 3. 90	Microwave calcination (800 °C, 1h, 200 °C × min ⁻¹)	99.5	Charge specific capacity of 354.1 mAh·g ⁻¹ at 0.1C and initial coulombic efficiency of 83.4%. Capacity retention rate 98.3% after 60 cycles at 0.1C	[74]
98.4	LIB was manually disassembled and obtained SG was leached with H_2SO_4 (200 g/l)	5:1	240	95	Heat treatment (900 °C), Ar 300 ml/min, 2 h	99.98	Reversible capacity of 358.8 mAh·g ⁻¹ at 0.1C and capacity retention of 95.8% after 100 cycles	[71]
34.2	LIB pretreated (dismantled and sieved etc.) and then obtained SG was leached with H_2SO_4 (0.5 M) and H_2O_2 (6%)	50 g/l	60	80	–	97.62	Initial discharge-specific capacity and coulombic efficiency at 0.2C 286.9 mAh·g ⁻¹ and 71.0%	[73]
–	LIB was manually disassembled, and obtained SG was 1&2) calcined, 3) leached with citric acid (0.2 M)	3. 1:50 g/ml	1. 60 2. 60 3. 50	1. 450 2. 500 3. 90	–	–	Discharge capacity of 330 mAh·g ⁻¹ after 80 cycles at 0.5C	[79]
76.0	SG anode was cut 2x2 cm, then leached with $(NH_4)_2S_2O_8$ (0.8 M)	60:1 g/l	60	80	–	–	Reversible capacity of 365.3 mAh·g ⁻¹ after 100 cycles at 0.1C and 330.2 mAh·g ⁻¹ after 500 cycles at 1C	[80]
96.8	1. SG mixed with 98.3% H_2SO_4 (1:1) + di-water (20% of total mass), 2. Roasted, 3. Leached with H_2SO_4 (200 g/l)	3. 7.5:1 ml/g	1. 20 2. 24 3. 60	2. 200 3. 90	High temperature calcination (1500 °C, 2 h, Ar atmosphere)	99.6	The initial charge capacity and retention rate are 349 mAh·g ⁻¹ and 98.8% at a rate of 0.1C	[70]

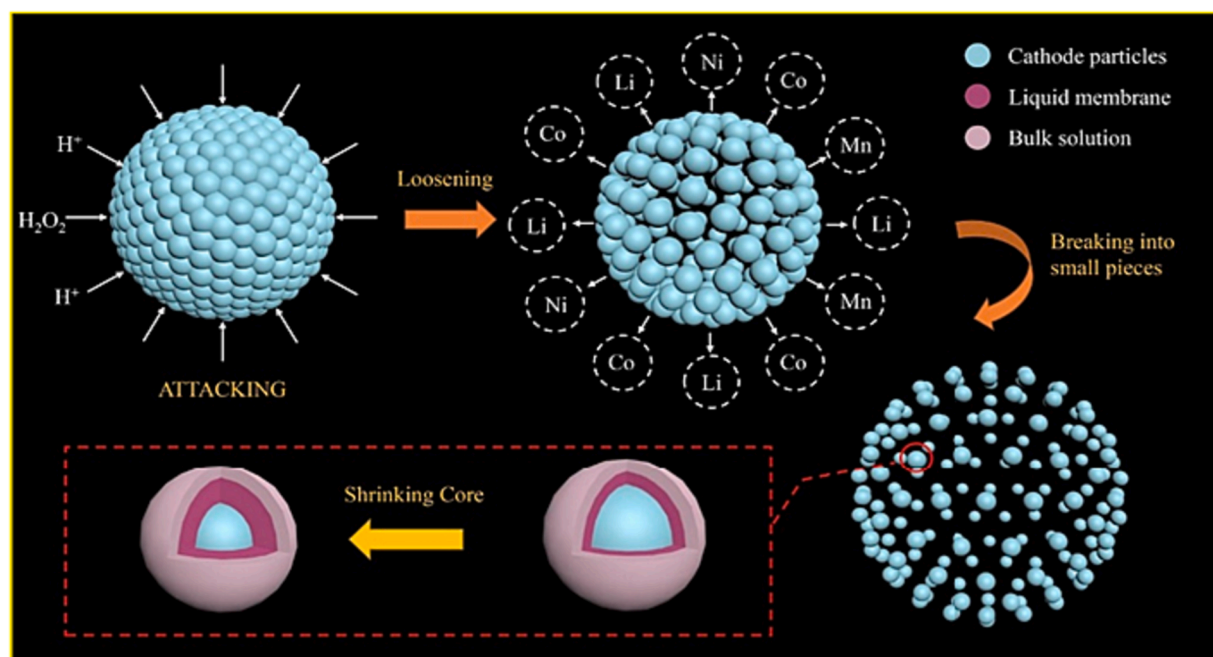


Fig. 8. A schematic representation of the leaching process's reaction mechanism. Reprinted with permission from Ref. [89]; Copyright © 2017 Elsevier.

used ozone without a neutralization agent to remove manganese from an NCM solution. The vast majority of the manganese (initial concentration of 1.26 g/L) was removed during the first 300 min of the reaction, but complete removal without neutralization required an additional 200 min[111].

After transition metal recovery, Li is still commonly present in sulfate solution. Various precipitation methods exist for the recovery of Li from wastewater from the metal sulfate recovery process. Li_3PO_4 and Li_2CO_3 [93] are the most common lithium compounds with low solubility and are separable from sodium. Wang et al. used a saturated solution of sodium carbonate to recover Li from a $\text{Li}_2\text{SO}_4/\text{Na}_2\text{SO}_4$ solution at a precipitation temperature close to 100 °C. The purity of the obtained precipitate was 96.97%. The drawback of lithium carbonate precipitation is the need for a very high precipitation temperature and residual concentration even after precipitation at 100 °C[93]. Li_3PO_4 has significantly lower solubility compared to Li_2CO_3 [112].

6.3.3. Solvent extraction

Solvent extraction-based techniques are effective for the treatment of spent LIBs because they can be operated continuously and require relatively simple equipment. In addition, relatively small quantities of reagents are needed, and they can achieve a high leachate throughput [113]. Several metal extractants have been reported (e.g., commercially available extractants Cyanex 272[114], PC88A[115], and D2EHPA [116]). The use of a very hazardous diluent, such as kerosene, is the main drawback of these extractants. In addition, multiple extractants are often needed to selectively recover valuable metals[117,118].

Ionic liquids (ILs) are organic salts, which are alternative, sustainable, and environmentally friendly extractants for the selective recovery of metals[118]. They typically have low volatility, vapor pressure, and flammability and are in a liquid form below 100 °C[119]. ILs consist of an organic cation and an inorganic anion. By choosing different cations and anions, it is possible to transform the properties of ILs. The cations and anions of ILs change during the solvent extraction process, which causes certain variations in the behaviors of metals[120]. This affects the mechanism and efficiency of extraction[51,121]. In addition, operating parameters, such as pH value, temperature, and extractant concentration, influence the extraction of metals. In the literature, many ILs for the extraction of metals from leaching solutions have been reported

[51,122,123]. Table 3 presents a few studies on the extraction and recovery of metals using different ILs[118,124,125,126].

The black mass obtained from LIBs recycling is commonly leached using sulfuric acid and a reducing agent. This results in a pregnant leaching solution (PLS) containing various amounts of Li, Ni, Co, Mn, and impurities, such as Al, Cu, and Fe. There are very robust precipitation methods for the removal of impurities, but at the very least, the separation of Ni and Co requires solvent extraction or ion exchange. Jantunen et al. studied the adjustment of synthetic NCM PLS representing old battery chemistries to NCM mixtures suitable for NCM 811 co-precipitation[127]. Manganese can be efficiently separated from Li, Ni, and Co using 0.8 M D2EHPA as the extractant. Manganese extraction amounting to 94.2% was achieved.

Similarly, Jantunen et al. used 0.8 M Cyanex 272 to recover the remaining manganese and 99.8% of the Co in three counter-current stages operating at pH 3.7–5.4. After Co removal, 98.1% of the Ni could be extracted using pre-neutralized 0.8 M D2EHPA in two counter-current stages. The operational pH was 7.2–7.7[127]. As demonstrated in earlier examples, D2EHPA is very suitable for selectively separating Mn from NCM leaching solutions. D2EHPA has great selectivity for manganese over Co and Ni. However, Cyanex 272 offers great selectivity for Co over Ni. Therefore, it is used industrially to separate Co and Ni from each other[127]. In another study, Lewatit TP260 (amino-methylphosphonic acid) ion exchange resin was employed to remove Al, Fe, Mn, and Cu from Co-rich leaching solution with approximately 1 % Co loss[128]. The aim of the study was to leave valuable Co, Ni, and Li in the raffinate. The results suggested that the purity of the Co-, Ni-, and Li-containing solution was over 99.6%. Mn and Cu could be eluted into a 2 M H_2SO_4 solution. The drawback of the treatment was that Al and Fe could not be eluted to sulfuric acid and required more expensive oxalic acid for recovery from the resin[128].

Even though ILs have many advantages, they also have some drawbacks such as high viscosity that makes them difficult to dissolve and diffuse and low contact area. Furthermore, it is difficult to separate and recover ILs when used in large quantities. It is possible to solve these drawbacks by immobilizing ILs on solid supports, these are called supported ionic liquids (SILs). SILs have the benefits of both ILs and solid support materials, including the ability to employ less IL, shorter diffusion distances, higher interface area, and increased transport rates.

Table 2

Acid leaching methods to recover different valuables from LIBs by using inorganic acid and organic acids.

INORGANIC ACID AS LEACHING AGENT					
leaching agent	Recovered metal(s)	Conditions	Recovered rate	Comments	Reference
1 M HNO ₃ + 1.0 vol-% H ₂ O ₂	Co and Li	Time: 1 h; Temperature 80 °C; S/L ratio: 20 g/L	100 % Co and Li	LIBs as raw material for leaching	[90]
1 M HNO ₃ + 1.7 vol-% H ₂ O ₂	Co and Li	Time: 1 h; Temperature 75 °C; S/L ratio: 20 g/L	95 % Co and Li	LIBs as raw material for leaching	[91]
3 M HCl + 3.5 % H ₂ O ₂	Co and Li	Time: 1 h; Temperature 80 °C; S/L ratio: 50 g/L	95% Co and 93% Li recovery after precipitation	LIBs	[92]
4 M HCl	Li, Mn, Co, and Ni	Time: 1 h; Temperature 80 °C; S/L ratio: 20 g/L	96.9% Li, 98.23% Mn, 96.94% Co and 97.43% Ni (expressed as purity-%)	LIBs	[93]
1 M sulfuric acid	Co and Li	Time: 4 h; Temperature 95 °C; S/L ratio: 50 g/L	Co 66.2%, Li 93.4% (expressed as efficiency-%)	LIBs	[94]
0.3 M sulfuric acid + H ₂ O ₂	Li and FePO ₄	Time: 2 h; Temperature: 60 °C; S/L ratio: H ₂ SO ₄ /Li molar ratio 0.57	Li 95.7 %, Fe < 1 %	LFP	[95]
4 M sulfuric acid + 10 vol-% H ₂ O ₂	Co and Li	Time: 2 h; Temperature 85 °C; S/L ratio: 1/10 g/L	Co 95% and Li 96% (expressed as efficiency-%)	LIBs	[96]
3 M sulfuric acid and 0.25 M Na ₂ S ₂ O ₃	Co, Cu and Co	Time: 3 h; Temperature 90 °C; Pulp density: 67 g/L	99.95 % Co, 99.71 % Li	Spent LIBs as raw material for leaching	[97]
2 M sulfuric acid + 15 vol-% H ₂ O ₂	Co and Li	Time: 0.167 h; Temperature 75 °C; Pulp density: 50 g/L	95 % Co and 100 % Li	LIBs waste as raw materials for leaching	[98]
ORGANIC ACIDS AS LEACHING AGENT					
Leaching agent	Recovered metal(s)	Conditions	Recovered rate	Comments	Reference
1.5 M lactic acid (LA)	Ni, Co, Mn, Li	Time: 20 min, Temperature: 70 °C S/L ratio: 20 g/L, [H ₂ O ₂]: 0.5 vol%	>98% Ni, Co, Mn, Li	After acid leaching of metals cathode material was resynthesized using sol-gel method	[99]
0.1 M citric + 0.02 M ascorbic acids	Li, Co	Time: 6h, Temperature: 80 °C	100 % Li, 80 % Co	Spent LIBs as raw material	[100]
2 M citric acid + 2 vol% H ₂ O ₂	Ni, Co, Mn, Li	Time: 90 min, Temperature: 80 °C S/L ratio: 30 mg/L	97 % Ni, 95 % Co, 94 % Mn, 99% Li	Spent LIBs as raw material	[101]
1.5 M succinic acid + 4 vol% H ₂ O ₂	Co, Li	Time: 40 min, Temperature: 70 °C S/L ratio 15 g/L	100 % Co, 96 % Li	Spent LIBs as raw material	[102]
0.5 M glycine + 0.02 M ascorbic acid	Co	Time: 360 min, Temperature: 80 °C, S/L ratio 2 g/l	95 % Co	Spent LIBs as raw material for leaching	[103]
0.5 M oxalic acid	Co, Li	Time: 150 min, Temperature: 95 °C, S/L ratio 15 g/L	Li 98 %, Co 97 %	Spent LIBs as raw material for leaching	[104]
2 M L-Tartaric acid + 4 vol% H ₂ O ₂	Li, Ni, Co, Mn	Time: 30 min, Temperature: 70 °C S/L ratio 17 g/L	99 % Li, Ni, Co, Mn	Spent LIBs as raw material for leaching	[105]
2 M formic acid + 6 vol% H ₂ O ₂	Al, Li, Ni, Co, Mn	Time: 50–120 min Temperature: 30–90 °C, S/L ratio 50 g/L	Al: 98%, Li: 98%, Ni, Co, Mn 100%,	Spent LIBs as raw material for leaching	[106]
1.5 M DL-malic acid + 2 % H ₂ O ₂	Co and Li	Time: 0.67 h; Temperature 90 °C; S/L ratio: 20 g/L	90 % Co and Li	Spent LIBs as raw material for leaching	[107]
1.25 M citric acid + 1 % H ₂ O ₂	Co and Li	Time: 0.25 h; Temperature 90 °C; S/L ratio: 20 g/L	> 90 % Co and 100 % Li	Spent LIBs as raw material for leaching	[108]
1.25 M ascorbic acid	Co and Li	Time: 0.33 h; Temperature 70 °C; S/L ratio: 25 g/L	94.8 % Co and 98.5 % Li	Cylinder spent LIBs of mobile phone as raw material for leaching	[109]

S/L ratio: solid/liquid.

Physical and chemical methods can be used to immobilize or support ILs on solid supports, and the supporting technique and support type have a significant impact on the SILs' properties [129,130]. The physical immobilization of an IL can be conducted for example through impregnation [131], sol-gel method [132], or encapsulation [133]. In the case of chemical immobilization, ILs are supported on solid supports via covalent bonds [134]. ILs can be immobilized in different materials such as carbon materials [135,136], silica [137], zeolites [138] polystyrene resins [139], polymer membranes [140], and metal-organic frameworks [141]. In literature, there exist many studies related to use immobilized ILs in extraction of metals [129,142–144]. Immobilized ILs have also been employed in research to recover metals from used LIBs [145]. As an illustration, Wang et al. (2022) created polymer inclusion membrane electrodialysis (PIMED) to separate Co(II) and Li(I) from the leaching solution used to extract used LIBs and findings revealed that solution purity was greater than 99.9% and 99.1% for Co(II) and Li(I), respectively. Zante et al. investigated selective separation of lithium ions from

complex aqueous solutions comprising sodium, cobalt and nickel ions by using supported liquid membranes (SLMs) impregnated with a mixture of IL 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([C₄mim][NTf₂]) where tri-n-butyl phosphate (TBP) was employed as the carrier [146].

6.3.4. Biohydrometallurgy of spent LIBs for metal recovery

Utilizing bioleaching (biohydrometallurgy) to recycle spent LIBs waste prevents precious elements from being lost to landfilling. The method relies on the metabolites that are generated during the microbial activity of microorganisms, which aid in the leaching of the metals from the LIBs waste [147,148]. For the bioleaching of metals from used battery trash, microorganisms that produce oxidizing agents (e.g., Fe³⁺), chelating agents (e.g., CN⁻), and organic and inorganic acids are usually employed [147,149]. Bacteria and fungi for metal recovery from LIBs waste have both been examined [150,151]. Species such as *Acidithiobacillus thiooxidans*, *Acidithiobacillus ferrooxidans*, *Thiobacillus*

Table 3
Metal recovery by using different ionic liquids.

Ionic liquid	Recovered metal(s)	Recovered rate	Comments	Reference
A fatty-acid-based ionic liquid	Co, Ni, Mn and Li	>99 % Co and 99 % Mn (two-stage), 100 % Ni and 100 % Li	The subsequent separation of Mn from Co: Extraction at 8 M HCl (pH = -0.9). Composite solution: ammonia, ammonium carbonate and ammonium sulphate. Co remains in the raffinate and Mn precipitates as MnCO_3 . The extraction of Ni: pH 1 and using a Na_2CO_3 solution. Li was separated independently from the organic acid used for the leaching procedure and adding EDTA to the aqueous phase	[118]
The 3-methyl-1-octylimidazolium thenoyltrifluoroacetone, Omim-TTA	Li, Co	>70 % of Li separating it from Co	Separation process: Mn: TODGA extractant diluted in an imidazolium-based IL Co: A phosphonium-based ionic liquid Ni: A DES (carboxylic acids and lidocaine) Li: e.g. chemical precipitation	[124]
An imidazolium-based and a phosphonium-based ionic liquid	Mn, Co, Ni, Li	$\approx 99\%$ Mn extracted in one step (pH = 3.3) SFMn/Co = 40–60, SFMn/Ni = 270–370 SFMn/Li = 500–700		[126]
A carboxyl-functionalized ionic liquid	Li	After the five-stage extraction: 96.80% of Li, 7.00% of Ni, 4.20% of Co, 13.6% of Mn	Organic phase: 80.0% TBP and 20.0% carboxyl based ILs with an O/A ratio of 2:1	[125]
A functional imidazolium ionic liquid	Li, Ni, Mn, Co	After a three-stage extraction: 92.5% of Li, <4.0 % of Ni, Mn, Co	Organic phase: 20.0%(vol) [HO2CMMIm] [NTf2] and 80.0%(vol) TBP with an O/A = 2:1, room temperature, initial pH of 5.0.	[121]

thiooxidans, *Sulfobacillus thermosulfidooxidans*, and *Aspergillus niger* have been explored in spent LIBs treatment[152]. Being able to thrive in a variety of pH ranges, tolerate toxic compounds, and operate at faster leaching rates, fungi offer advantages over bacteria[153]. Chemotrophic bacteria can be employed for metal dissolution from spent LIBs. Oxidation of ferrous to ferric ions and elemental sulfur to H_2SO_4 in the process of dissolution provides energy for chemotrophic bacteria. In the bioleaching process, chemolithotrophic microorganisms produce sulfuric acid and ferric ions for proton and oxidative attacks on waste

materials, respectively, for metal leaching. Fungi and heterotrophic bacteria produce many metabolites, including organic acids and surfactants, by utilizing organic carbon[154,155].

A. niger fungal species produce organic acid, which can dissolve metal from spent LIBs[147,156]. Depending on the mechanism and operational basis, the bioleaching processes can be differentiated into acidolysis, redoxolysis, or complexolysis(Fig. 9)[51,149]. With the aid of protons or acids, insoluble metal species are transformed into soluble forms in acidolysis. The dissociation of biogenic acids produces protons,

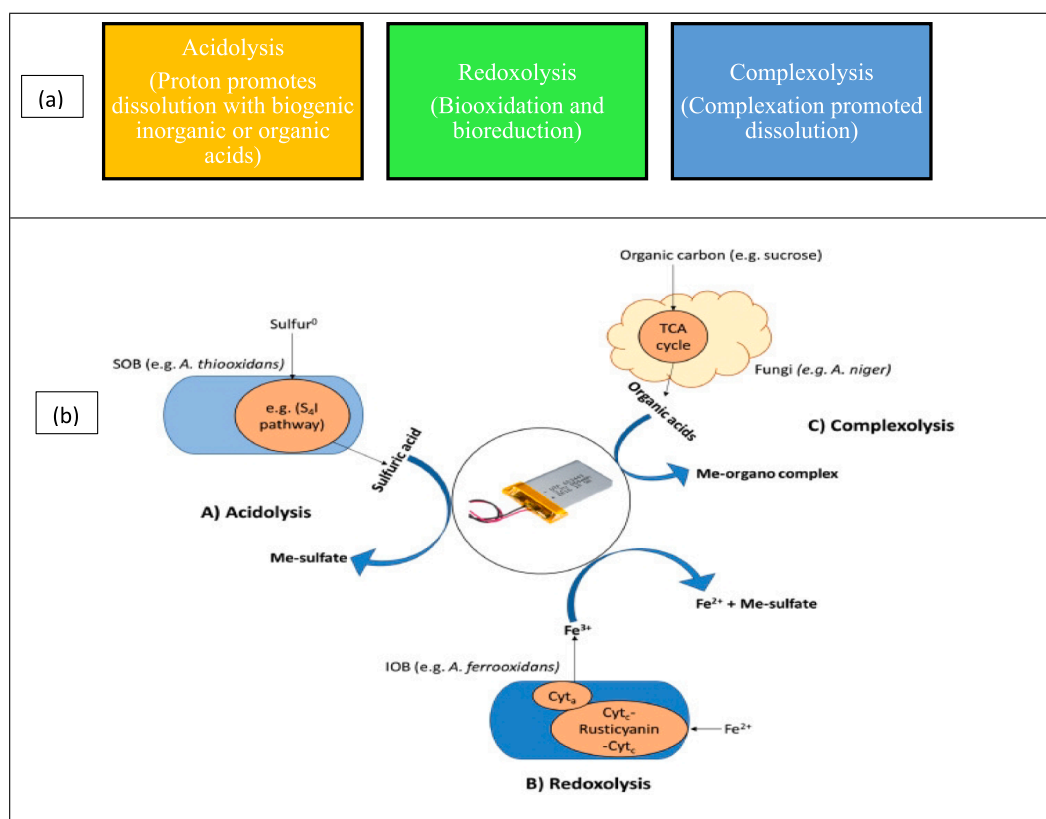


Fig. 9. (a) Different bioleaching processes (b) Microbial activity for metal solubilization (TCA cycle –tricarboxylic acid cycle, Cyt– cytochrome, S_4I pathway–tetrathionate intermediate thiosulfate oxidation pathway). Reproduced with permission from Ref. [149], Copyright © 2020 Elsevier.

which eventually target the oxygen on the surfaces of metals to trigger metal dissolution. Bacteria typically use extracellular polymers (EPS) and biofilm growth in redoxolysis to adhere to the surface of minerals. By transferring electrons from the solid substrate to the microorganisms, the metals are subsequently brought into the solution. Occasionally, during the bioleaching process, both acidolysis and redoxolysis can occur. Wu et al. showed that the proximate cause of the Co dissolution from LIBs is redoxolysis bioleaching[157]. In the complexolysis process, a soluble metal–organic complex is created during the chelation reaction between the metal ions and the secondary metabolites generated by the bacteria[158]. Interestingly, complexolysis controlled the fungus-based bioleaching of Co and Li from LIBs[156]. Fungi and heterotrophic bacteria can produce a range of organic acids, such as malic, citric, and oxalic acid. These biogenic organic ligands can also generate soluble metal–organic complexes by chelating with metal ions.

The biohydrometallurgical approach has been extensively investigated for metal recovery from spent LIBs[29,30,152,154,159–162]. Niu et al. explored the bioleaching of spent LIBs by employing a mixed culture consisting of *Alicyclobacillus* sp. and *Sulfobacillus* sp.[163]. A bio-electro-hydrometallurgical approach successfully recovered 91.45%, 93.64%, and 87.92% of Mn, Co, and Li, respectively, from cathode active materials[164]. To recycle Co from LiCoO_2 , a copper-catalyzed bioleaching with *A. ferrooxidans* was accomplished[165]. Noticeably, only 43.1% of the cobalt dissolved in the absence of Cu ions. Various factors, such as the availability of growth nutrients, pulp density, pH, nature/type of microbial species, and concentration of carbon sources, can affect bioleaching efficiency[34,52,147,166].

The growth medium parameters that eventually affect the dissolution of metals from LIBs rely on the quantity of organic acids produced by microbial species. Hence, parameter optimization is essential in the improvement of the recovery of resources from used LIBs. The nature and quantity of carbon sources can impact microbial growth, which in turn affects organic acid production[149,167]. The effective recovery of Cu (100%), Li (95%), Mn (70%), and Al (65%) metal was achieved when *A. niger* was employed at a pulp density of 1%; however, only 45% and 38% efficiency was achieved for Co and Ni, respectively[156].

To bioleach metals from used coin cells, Naseri et al. examined *A. ferrooxidans*, and the pulp density of 40 g/L showed the maximum leaching efficiency (100% for Li, 88% for Co, and 20% for Mn) in 12 days[168]. In another study, *A. niger* was employed for Li, Mn, Cu, Al, Co, and Ni recovery from spent LIBs from mobile phones, and the impact of different parameters was optimized by applying the response surface methodology[147]. Li, Co, Mn, and Ni were recovered from spent LIB cathode materials with a 1% pulp density of *A. thiooxidans*, and *Leptospirillum ferriphilum* resulted in approximately 95% of metal leaching[169].

In another investigation, the bioleaching of spent NMC-based LIBs was assessed using *A. ferrooxidans*[152]. Biswal et al. employed *A. niger* strains to recover Co and Li[160]. Similar to pulp density, substrate concentration also impacts bioleaching, and different microbes can perform differently with different substrates. Temperature can also impact bioleaching[165,167,170]. Temperature ranges from 30 °C to 35 °C have been widely used in the bioleaching of metals with *A. ferrooxidans*[165,167]. In the case of fungal-based bioleaching using *A. niger*, 30 °C was also suitable for metal leaching from LIBs[156,159]. Additionally, the cathode material (LiCoO_2) of spent LIBs was treated with *A. ferrooxidans* to recover precious metals[171]. The presence of higher Fe^{2+} concentrations resulted in a reduction in dissolution as a result of Fe^{3+} co-precipitating with the metals in the residues. Fan et al. employed *A. niger*, as it can produce glucose oxidase, which can produce gluconic acid via glucose oxidation[161].

Wu et al. claimed that while the bioleaching of Co is dependent on the quantity of Fe^{2+} ions, increasing the supply of sulfur enhanced Li bioleaching from LIBs[157]. *A. ferrooxidans* and *A. thiooxidans* were used by Heydarian et al. to examine the bioleaching of LIBs, and the response surface technique was applied to optimize variables, such as

initial pH, concentration of iron sulfate, and sulfur content[172]. Oxidation reduction and acid dissolution procedures facilitated the recovery of Li, Ni, and Co. In bioleaching, pH is an important factor that can affect the growth and activity of microbial species, which have a direct impact on the production of secondary metabolites and ultimately affect the bioleaching efficiency[147]. Iron-oxy hydroxides can precipitate in the case of acidophilic bioleaching at higher pH and greater ferric ion concentrations, which can form a passive layer on the surfaces of leachable substrates and therefore hinder the bioleaching process[152].

The choice of microbial species is the first and most important step in the design of the bioleaching processes in biohydrometallurgy, which is dependent on a number of factors, such as the material's composition, the target metals, and even the downstream process. Owing to its potential environmental benefits in metal recovery, and the fact that it is eco-friendly and safe to handle, it offers low energy consumption, minimizes waste generated, and consequently reduces treatment costs, bioleaching/biohydrometallurgy has gained the interest of many researchers[29,152,173]. However, bioleaching is still only implemented on a small scale in laboratories due to constraints, such as delayed kinetics, poor leaching effectiveness, and low pulp density[15].

6.3.5. Other methods for resource recovery from LIBs

Membrane-based and electrochemical methods have also been studied recently in conjunction with other recycling methods. For recovering metal from LIBs waste, membrane technology could represent an appealing approach as it offers a sustainable and effective solution to separate the metal ions found in the waste stream. In accordance with the particular metal ions of interest and their concentration levels, various types of membranes, such as ion exchange membranes or nanofiltration membranes, can be used. Target metal ions preferentially pass through the membrane while impurities and other ions are retained. Furthermore, a membrane-based hybrid system was employed for metal recovery from acidic leachate of spent LIBs cathode material[38]. After the initial alkaline pretreatment of battery leachate, Fe and Al was successfully separated using the hybrid membrane system by bringing the pH down to 5.53, and after that Li was separated from the bivalent metal ions using UF and NF membrane filtration. In another study, the recovery of Li from spent LIBs was examined by employing a novel ion-imprinted membrane (SP-IIM)[174]. The SP-IIM's maximum capacity for Li(I) adsorption was $42.58 \text{ mg} \cdot \text{g}^{-1}$. The SP-IIM's adsorption capacity only declined by 4.6% after five consecutive adsorption–desorption cycles, demonstrating the SP-IIM's remarkable reusability. Another study explored the hybrid electrobaromembrane (EBM) approach for separating the Li(I), Co(II) and Ni(II) from spent LIBs leachate[175]. The efficiency and effectiveness of metal recovery from LIBs leachate by using membrane-based technology depend on numerous factors including membrane material, pore size, operating conditions, and the composition of the LIBs waste.

Another promising way of recycling LIBs is through electrochemical recycling approach (ERA). It is widely employed to recover metal elements such as gold, silver, copper, and nickel in industrial wastewater[176], valuable elements in tailings[177], and lithium from salt lake brine[178]. This method has huge potential for large-scale application in LIBs recycling because it can have high selectivity to recover different ions by only controlling the voltage, current, and conditions to successfully separate and recycle the desired elements. When compared to other processes, ERA consumes less energy and reagents than hydrometallurgical procedures and pyrometallurgical processes. Consequently, it has significant development potential and is efficient and environmentally beneficial[179]. As a pre-treatment phase, ERA can be used to separate the current collector from the active material/graphite. In this method, the utilized electrodes serve as working electrodes, and the current supplied to them strips the active material from the current collector[180]. In addition, for cathode recycling, which represents the great difficulty in recycling battery components by ERA, a leaching step

is needed, in which the separation efficiency is determined by the applied current instead of traditional chemical reagents, which greatly diminishes the chemical's consumption and less amount of wastewaters is generated and with less toxicity[180]. Although ERA is a very promising strategy for battery recycling it also possesses important issues that need to be addressed, for instance, challenges in preventing the production of side reactions during the electrolysis process, which increases the power consumption[36]. Indeed, the ERA although very promising it is still immature for cathode materials and this area needs to undergo additional research and development.

7. Current industrial-scale LIBs recycling approaches

Lithium can be produced from several kind of naturally available sources like different minerals (spodumene, for example), clays (hectorite, for example) and salt lakes[181] and it is reported that 250 tonnes of ore or 750 tonnes of brine are needed to produce 1 tonne of virgin lithium[36,182]. Lithium can be extracted from mineral by using acid or chemical treatment. These chemical processes may include also chlorination part[183]. It was reported that whole-battery GHG emissions might be as much as 50% lower when batteries employed recycled cathode, aluminium, and copper as contrasted to batteries utilizing solely virgin materials when a closed-loop scenario on the operation of battery recycling was explored[184]. Recycling battery components can be economical considering the high cost of producing batteries. In 2019, it was anticipated that 500,000 tonnes of batteries will be produced worldwide; of them, 15,000 tonnes of Al, 60,000 tonnes of Co, and 75,000 tonnes of Li could be recycled[184–186]. In addition of those, spent LIBs are also the well available secondary resource for lithium and other metals. Processes are also chemical-intensive including hydrometallurgical processes combined with pyrometallurgical treatment. In pyrometallurgical processes the main disadvantage is the lack of lithium recovery. The advantage of hydrometallurgical processes is the recovery potential of each metals [187]. However, complete recovery of material from spent LIBs waste is still challenging. Various extraction techniques,

including pyrometallurgy, hydrometallurgy, or a mixture of the two, have been employed in industrial recycling operations by various industries[9,14,20]. Table 4 is a list of the various industries that deal with recycling LIBs[188189190191].

As highlighted previously, there are three main approaches to battery recycling: pyrometallurgical, hydrometallurgical, and direct recycling processes. Among these, pyrometallurgical and hydrometallurgical approaches are popularly employed at the industrial level, while the third is being explored only at the laboratory level, which means that much future effort is required to scale such methods for commercialization. The first two methods rely on two steps: the destruction of the crystal structure of cathodes to the atomic level and the extraction of the desired metals. The most notable disadvantages of pyrometallurgy are related to the formation of harmful or toxic gases and to the high energy demand. Therefore, the most appropriate recycling procedures are based on hydrometallurgical processes. The main advantages and disadvantages of LIBs recycling are summarized in Table 5.

8. Challenges in recycling spent LIBs

Leaching agents, such as HCl, H₃PO₄, or H₂SO₄, and oxidative agents, such as H₂O₂, are used in the LIBs recycling process[14,193–195]. LIBs recycling can have negative consequences for the environment due to the production of side streams and harmful compounds [16,22]. A variety of organic solvents are also utilized in the recycling process[4,9,20,196]. Sulfuric-based LIBs leaching has negative environmental impacts, such as ozone depletion, freshwater eutrophication, and human toxicity[197]. Fluorine compounds, hazardous organic compounds, greenhouse gases, and chemicals are among the undesirable pollutants that recycling processes are known to produce in considerable quantities. Poor recycling of LIBs can have a significant detrimental effect. For example, improper disposal of toxic and flammable components, such as Ni, Co, and electrolytes, leads to explosions and results in soil and groundwater contamination. The treatment of exhaust gases and the recovery of chemical waste and wastewater produced during the

Table 4
LIBs recycling approaches in different countries.

Company	Country	Techology	Details	Reference
Accurec	Germany	Mech + Pyro + Hydro	Thermal pre-treatment, mechanical separation, pyrometallurgical treatment and hydrometallurgical recovery of Co- and Ni-salts.	[188]
AkkuSer Ltd.	Finland	Mechanical	Manual sorting followed by crushing, magnetic separation, and sieving. Separated fractions goes to various plants or smelters.	[188]
Batrec AG	Switzerland	Mech + Hydro	Shredding, mechanical separation and acid leaching followed by separation processes	[189]
Brunp	China	Thermal	Thermal pre-treatment, mechanical separation, pyrometallurgical treatment and hydrometallurgical recovery of NCM	[188]
Ganzhou Highpower	China	Mech + Pyro + Hydro	Mechanical processing, pyrometallurgical treatment and hydrometallurgical recovery of metals.	[188]
GEM	China	Mech + Hydro	Sorting, comminution and hydrometallurgical recovery of metals.	[188]
Glencore Plc	Switzerland	Pyro + Hydro	Treats small battery packs pyrometallurgically and matte goes to hydrometallurgical refining	[191]
Fortum Waste Solutions	Finland	Mech + Hydro	Mechanical processing in various locations. Hydrometallurgical sulfuric acid leaching and metal separation in Harjavalta, Finland.	[192] (ESAV I/18974 /2020)
Inmetco	USA	Pyro	Calcination and alloying.	[191]
Nickelhütte Aue	Germany	Pyro + Hydro	Thermal pre-treatment, pyro-treatment (NiCoCu-matte + slag) and hydrometallurgical separation of Ni, Co & Cu.	[188]
Ontoprocess	USA	Mech	Mechanical separation and electrolyte extraction using supercritical CO ₂	[190]
Snam	France	Thermal + Pyro	Thermal pre-treatment, pyro-treatment and battery metals are sent for further processing.	[188]
Toxco	Canada	Mech	Mechanical treatment and Li recovery	[190]
Umicore	Belgium	Pyro + Hydro	Smelting, leaching and solvent extraction	[188]

Table 5

Comparison of main advantages and disadvantages over different LIBs recycling processes.

Process	Hydrometallurgy	Pyrometallurgy	Direct recycling
Advantages	Wide range of waste materials	Elevated recovery rate and high purity of valuable metals (e. g., Li)	Low cost, low consumption
	Short process flow	Low energy input	Less waste (solid and liquid) generated
	No need of pre-sorting	Reuse of the recycled materials for cathode fabrication	Less energy input
	Consolidated technology and widely employed	Consolidated technology and widely employed	
Disadvantages	Insufficient efficiency on Li and Al removal	Need of pre-sorting	Extensive pre-sorting/separation processes
	High gas generation and high cost for treating it	More complex for designing the process	Applied to a small (lab) scale
	Low selectivity of recycling elements	Large usage of chemicals	
	High energy inputs	Waste generation (liquid effluents)	

recycling process incur additional expenses. The physical and chemical characteristics of electrolyte components make recycling more difficult, and it is difficult to separate and collect electrolytes because they can get stuck on the electrodes during recycling[9]. During treatment, very harsh conditions, such as strong acid, high temperature, and lengthy subsequent extraction processes, are usually required for cathode materials due to their stable structures[32]. The possibility of the entrapment of metals and graphite by the organic compounds utilized in extraction reduces the effectiveness of the metals' and graphite's separation.

During thermal treatment of graphite carbon materials, gases such as CO and CO₂ can be formed in the presence of strong oxidants[14]. Estimating the right amount of CO₂ emissions in battery production and recycling is not a simple task because it involves a series of steps, materials, and processes. Recycling LIBs culminate in saving important materials that are necessary for the long-term sustainability of LIBs and for the reduction of CO₂ emissions. The manufacturing of cathodes and the battery assembly process are the two processes that produce the majority of the 91.21 kg CO₂-eq/kWh, of carbon emissions from the battery industry[198]. Comparatively, the CO₂ emissions of batteries made from recycled materials are 51.8% lower than those of batteries made from raw materials[199]. Depending on the chemistry and which used materials the CO₂ emissions can vary, with cobalt and nickel being important elements for increasing the emissions. For instance, compared to LIBs with NMC, LFP batteries emit roughly 42.0–44.5% less CO₂, and their ecological and water footprints are also lower[198]. Depending on the materials used, how they are produced, and the energy sources employed in the operations, different amounts of CO₂ are released during battery manufacture and recycling. Then, a deep study focused on the LCA analysis of battery recycling scoping the CO₂ emissions in each step of the recycling is necessary to elucidate important aspects of CO₂ footprint in battery manufacturing and recycling.

Recycling valuable resources from used LIBs waste for reuse is a sensible strategy owing to the increasing demand for LIBs. When establishing battery recycling strategies, a number of factors, such as energy usage, GHG emissions, material flow, and industrial aspects, must be taken into account. To assess the overall energy consumption aspect of critical minerals recycling from LIBs, all stages of the supply chain should be considered. Energy is required in all steps involved in

the recycling process and energy consumption in the treatment of spent LIBs waste can vary depending on the recycling processes, technologies employed, and the condition and composition of the spent batteries being treated. Although pyrometallurgical processes are generally straightforward to execute, they have high energy and environmental costs for burning and calcining the batteries[36]. Despite hydrometallurgical processes use less energy than pyrometallurgical processes during processing, multiple chemicals are required, and water used during process needs to be cleaned thereafter. For electrochemical recovery process also consume energy depending on the current density applied, and the efficiency of the electrodes. When metals, electrolytes, and graphite, are recycled in a closed-loop recycling scenario, a reduction in overall energy consumption may be possible [200]. Continuous efforts are being made to optimize recycling processes, develop more energy-efficient methods, and implement sustainable practices to reduce the energy consumption and environmental impact of metal recovery from spent LIBs.

In addition to the production of toxic compounds and solvents, LIBs recycling presents a number of other challenges:

- (i) **Cost:** When compared to conventional waste disposal techniques, recycling LIBs can be more expensive, as they require specialized facilities, processes, and equipment. This can make recycling less accessible and limit the number of batteries recycled.
- (ii) **Collection and transportation:** Collecting and transporting used LIBs for recycling can be challenging and expensive, as batteries may need to be transported over long distances to reach a recycling facility.
- (iii) **Low recycling rate:** Regardless of the benefits of recycling, the recycling rate of LIBs remains low in many regions. Demand for industrial battery recycling will increase as a result of the new EU battery regulations.

It might be possible to minimize the negative consequences of spent LIBs on the environment and conserve valuable resources by improving recycling procedures, expanding the collection and transportation of spent batteries, and raising public awareness about the importance of recycling.

As previously pointed out, LIBs recycling involves several steps with highly complex processes, and with the continuous evolution of LIBs technology aiming to increase battery efficiency, new composition/components have been manufactured. Applying a standardized recycling technique is exceedingly challenging due to these differences in the LIBs composition, leading to reduced efficacy. These challenges eventually disappear with the expansion and standardization of the LIBs market.

9. Future perspectives

Prior to recovering valuable metals from spent LIBs, mechanical separation can be scaled up to increase recycling efficiency and decrease chemical reagent consumption. Due to the increased pace of waste production compared to existing recycling capacities, battery repurposing should be envisaged as an alternate end-of-life option. With the potential to maintain 70% to 80% of their prior capacity, retired batteries could be reused in smaller storage systems[201,202]. Pretreatment processes can also significantly increase the effectiveness of upstream recycling operations. The incorporation of different approaches, such as pyrometallurgy and hydrometallurgy, can improve recycling efficiency. Most studies have focused on recycling battery anode and cathode components, ignoring the recycling of electrolytes, which entails environmental problems[9]. In recent years, artificial intelligence (AI)-based approaches have received much attention; however, they are not currently deployed on a broad scale[12,16]. Future studies can employ AI-based approaches to provide more effective recycling approaches[203]. More attention ought to be focused on

preventing pollution at the source and recycling LIBs using environmentally benign reagents to reduce secondary contamination and achieve environmentally sustainable practices.

The future of spent LIBs recycling is very promising, as the LIBs demand continues to grow in response to the increasing demand for EVs and other battery-powered products. To conserve resources, it will be necessary to recycle more spent batteries as the number of LIBs in use escalates. It is anticipated that recycling rates will rise in tandem with continued growth in the volume of LIBs in use. This will be driven by a combination of regulatory and market forces, as well as consumer demand for more sustainable products. Examples of such forces are as follows:

Technological advancements: Advances in recycling technology are expected to improve the efficiency and cost-effectiveness of LIBs recycling, making it an increasingly attractive option for recovering valuable resources.

Growing demand for recycled materials: As the demand for LIBs continues to grow, so will the demand for recycled materials that can be used as feedstock in the production of new batteries. This is expected to drive the growth of the circular economy for batteries.

Regulatory support: Governments around the world are expected to continue to provide regulatory support for spent LIBs recycling, including requirements for the collection and recycling of spent batteries, as well as incentives for technological developments.

Finding more economic recycling processes: The majority of the economic benefits are acquired from the precious metals found in the final products, which serve as a direct incentive for manufacturers to recycle used LIBs. As a result, it is essential to lower the price of the entire recycling process.

Development of closed-loop recycling: As the volume of LIBs in use continues to grow, it is expected that more closed-loop recycling systems will be developed in which used batteries are recycled to produce new batteries in a sustainable and circular manner.

Overall, it is anticipated that LIBs recycling will significantly contribute to resource conservation, reduce the environmental impact of resource extraction, and advance the growth of a circular economy for batteries.

10. Conclusion

The production of LIBs has substantially increased as a consequence of the ongoing surge in demand for LIBs. The quantity of spent LIBs has been steadily rising as demand has grown, posing a difficult waste management challenge for recycling and placing an extensive burden on the resources required for battery production. Moreover, repurposing EV batteries could reduce the need for new battery production. The goal of LIBs recycling is to conserve valuable resources, reduce the harmful environmental consequences of resource extraction, and accelerate the growth of a circular economy for batteries. The recycling of LIBs requires specialized facilities, processes, and equipment, and it can be more expensive than traditional waste disposal methods. As the demand for LIBs continues to grow, recycling will play an increasingly important role in resource conservation and in reducing the environmental impact of resource exploitation.

Despite a substantial increase in the volume of spent LIBs, recycling technologies and innovations are not evolving at the same rate. This might be attributed to constraints in the technology and infrastructure involved in LIBs recycling processes, as well as to the inadequate collection of spent LIBs. Several approaches, such as pyrometallurgy, hydrometallurgy, and biohydrometallurgy, have been evaluated for resource recovery from spent LIBs. Research has shown that an optimal combination of these approaches may result in the most effective recycling method. Compared to pyrometallurgy, hydrometallurgy is a technology that is more economical and environmentally benign. Nevertheless, hydrometallurgy generates significant amounts of corrosive effluent that are difficult to recycle and reuse. Furthermore,

additional efforts may be required to recover materials for reuse in the recycling process. Although the biohydrometallurgy process has little impact on the environment, some drawbacks, such as a slow kinetic rate and limited tolerance for harmful chemicals, prevent it from being widely used. The selection of microorganisms in the biohydrometallurgy process is crucial, as it can have a significant impact on resource recovery efficiency. Prioritizing the development of abundantly available non-toxic raw materials for the production of LIBs would help in reducing environmental issues and enabling better LIBs treatment. Furthermore, finding substitute materials for the existing toxic components of LIBs and investigating inexpensive synthesis methods are crucial steps in lowering the environmental impact of LIBs. Consequently, to ensure the effective recycling of LIBs, multidisciplinary research is necessary.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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