



Treatment and recycling of spent lithium-based batteries: a review

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Abstract

Lithium-ion batteries (LIBs) have a wide range of applications from electronic products to electric mobility and space exploration rovers. This results in an increase in the demand for LIBs, driven primarily by the growth in the number of electric vehicles (EVs). This growing demand will eventually lead to large amounts of waste LIBs dumped into landfills which can cause serious environmental problems. To reduce this environmental impact, the need for efficient recycling technology emerges. Furthermore, it has been proven that recycling waste LIBs consume less material and energy than producing new ones from virgin materials. Therefore, this paper aims to review different recycling technologies including hydrometallurgy, pyrometallurgy, direct recycling, and precise separation. The review concludes that hydrometallurgy might be the most efficient method of recycling waste LIBs on an industrial scale.

Keywords LIB · Recycling · Hydrometallurgy · Pyrometallurgy · Precise separation

Abbreviations

ANVIL	Adhesion Neutralization via Incineration and Impact Liberation
CAM	Cathode active material
CTR	Carbothermic reduction
Cyanex 272	Bis(2,4,4-trimethyl-pentyl) phosphinic acid
D2EHPA	Di(2-ethylhexyl) phosphoric acid
DEC	Diethyl carbonate
DMC	Dimethyl carbonate
EC	Ethylene carbonate
ECS	Eddy current separator
EMC	Ethyl methyl carbonate
EV	Electric vehicle
GHG	Greenhouse gas
HTMR	High-temperature melting recovery
LCO	Lithium cobalt oxide
LFP	Lithium iron phosphate
LIB	Lithium-ion battery
LMO	Lithium manganese oxide
NCA	Nickel cobalt aluminum
NMC	Nickel manganese cobalt
NMP	N-methylpyrrolidone

PC	Propylene carbonate
PTFE	Polytetrafluoroethylene
PVDF	Polyvinylidene fluoride
SEI	Solid electrolyte interface
STF	Single tube furnace
TBP	Tributyl phosphate
TFA	Trifluoroacetate
UHT	Ultra-high-temperature smelting technology

Introduction

Recently, the demand for lithium-based battery-operated electronics, solar panels, e-scooters and, most importantly, electric vehicles (EVs), has increased. As a result, lithium-ion batteries (LIBs) are being consumed at an exponential rate and are becoming a necessary raw material for many industries. If used batteries are not treated properly, they will create a massive amount of environmental waste which would be difficult to handle. This has been the interest of environmentalists who have investigated different methods to recycle such LIBs and, thus, reduce their damage to the environment. Despite the different uses of LIBs, EVs have the most prominent demand because their carbon footprint over their entire lifetime is far less impactful on the environment than that of an engine-driven car. Additionally, unstable oil prices, relatively low maintenance costs and better performance are other factors that contributed to the

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increasing demand of EVs. To elaborate, about 2 million EVs were sold worldwide in 2019, with sales projected to hit 21 million by 2030 [1]. This demand is expected to continue growing to reach 60 million EVs by 2040 [2]. Furthermore, it is expected that the UAE will have around 42,000 EVs on the road by 2030 [3].

To manage the environmental waste from LIBs, a recycling-based circular economy is becoming a necessary option. In a circular economy, materials are reused, repurposed, or retrieved locally instead of being dumped in the landfill [4]. To address the issue of circular economy, the UAE has already joined the World Economic Forum's initiative, Scale 360. The UAE expects to house one of the most extensive waste-to-energy facilities in the world [5]. In recent years, notably during the Dubai Expo, there has been much discussion about the urgent need to build more sustainable lifestyles and a more circular economy. They are conscious of the environmental vulnerability, climate change, biodiversity loss, and plastic pollution that can pose imminent risks [5].

Furthermore, the “Energy Consultants Circular Energy Storage” analysis utilized data from more than 50 of the world's largest recycling companies to get the most recent numbers [6]. Their findings suggest that of the 180,000 tons of LIBs available for recycling in 2019, about 97,000 tons were recycled; 67,000 were processed in China, 18,000 tons in South Korea, and the remaining 12,000 tons were recycled in Europe, Japan, United States and Canada. However, the study does not mention any recycling done in the UAE or the MENA region. Additionally, those recycled LIBs can perform better than new ones since they are more porous. Higher porosity means larger surface area for the electrochemical reactions to occur at the electrodes, thus, faster

charging [7]. Therefore, to support the anticipation of the increased demand for EVs in the future, this review aims to study the treatment and recycling of the waste produced from LIBs in general, and those used in EVs in particular. A focus is also placed on the novel technologies that are being employed by various countries to recycle spent LIBs. These technologies can also be applicable to cobalt-based batteries. This effort assists in stimulating a circular economy within the batteries industry.

LIB considerations

LIB components

The LIB cell is composed of several layers which are enclosed together in a metallic shell or case, as shown in Fig. 1 [8]. These layers include five main components, all of which are recyclable and have the potential to be used again. Figure 1b shows these components, namely anode, cathode, electrolyte, separator, and two current collectors. The anode material mostly utilized is graphite, whereas the cathode used could be LiCoO_2 (LCO), LiMn_2O_4 (LMO), LiFePO_4 (LFP), and nickel–cobalt–aluminum (NCA) [9]. The cathode also contains an organic binder, such as polyvinylidene fluoride (PVDF) [8]. Lithium salts, such as LiPF_6 , LiBF_4 , and LiClO_4 , are combined with organic solvents, such as propylene carbonate (PC), ethylene carbonate (EC), ethyl methyl carbonate (EMC), dimethyl carbonate (DMC), and diethyl carbonate (DEC), to form electrolytes that serve as a conductive pathway for Li ions to move. A separator is placed between the cathode and the anode to prevent physical contact; it is made of polymeric membranes or

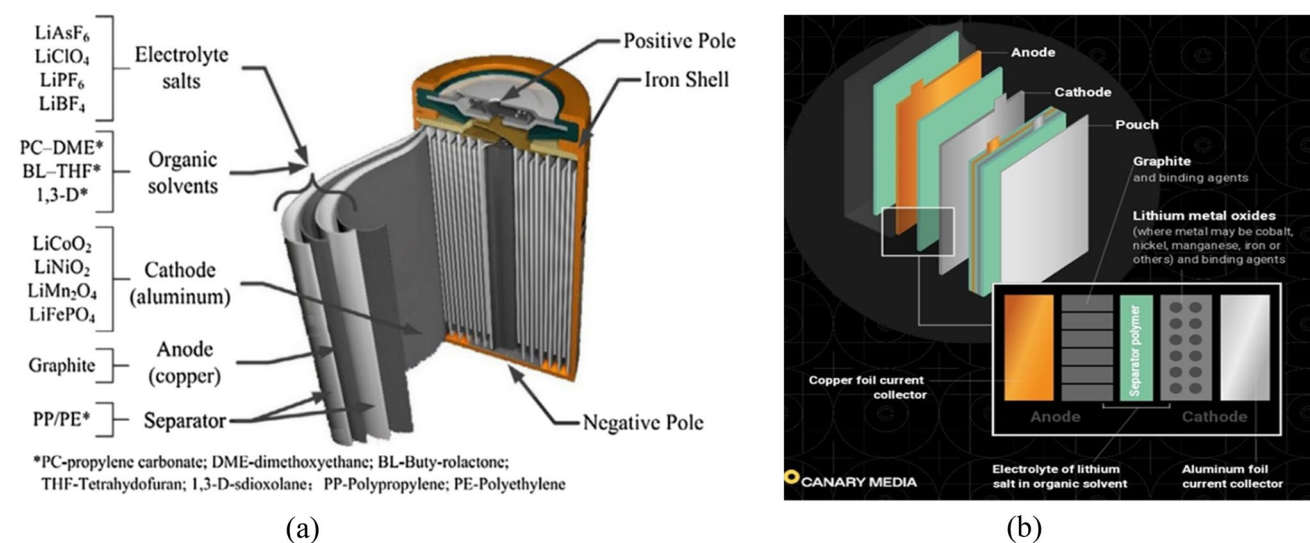


Fig. 1 Structure **a** and layered structure of LIB batteries **b** [8, 9]. Copyright 2021 Elsevier

non-woven fabric mats in layers. Furthermore, the electric current generated at the electrodes is collected by bridging elements called current collectors. Aluminum and copper foils are used as commercial current collectors for cathodes and anodes, respectively [8].

The weight percentages contribution of each component to the LIB is shown in Fig. 2; it is seen that the case, the cathode and the anode contribute largely to the LIB's weight by 30.2, 25.5, and 14.5%, respectively [10].

LIB cell construction

LIBs are available in 3 different constructions: cylindrical, prismatic, and pouch cells. The cylindrical cell is made by rolling the cathode, separator, and anode foil together, and enclosing them by a stainless steel or aluminum cell housing. Their round and small shape allows them to be stacked in devices of all sizes [11]. Cylindrical LIB cells are widely used in laptops, EVs like Tesla and Lucid, and even in the Mars exploration rover [12].

On the other hand, prismatic cells use a flat rectangular housing to lower the thickness of the cell. The cell components are either rolled like cylindrical cells and flattened or can come in rectangular stacks. This shape of batteries is common in electronics since they can be replaced easily. Additionally, these cells can shut off current flow out of the battery if the temperature becomes very high. They can also release internal pressure in case of gas accumulation inside the cell [11].

Lastly, pouch cells which are also known as lithium polymer, are similar in shape to prismatic cells, but they use a flexible polymer/aluminum housing instead of a rigid metal case. By eliminating the metal case, pouch cells save on cost, weight, and thickness. However, it does not offer the same

safety as prismatic cells since it can be prone to swelling [11]. Cell swelling occurs when the current keeps accumulating inside the cell, leading to buildup of heat and gas, which can pose a serious safety hazard [13]. Consequently, pouch cells are not preferred in EVs, but can be used in drones and smartwatches [14].

Charging/discharging cycles of LIBs

The schematic of the LIB cell, its charging and discharging cycles are displayed in Fig. 3. When batteries are discharged, Li ions move from the anode to the cathode, which creates current flow due to the generation of electrons. The opposite occurs when the batteries are charged where an external power is used [15]. The movement of Li ions occurs inside the cell with the help of the electrolyte, while the electric current flows in the external circuit and is measured by a voltmeter.

Furthermore, the charging or discharging rate of the battery is expressed in fractions or multiples of the C rate. For example, a C/2 charge or discharge rate means that the battery will be charged or discharged in two hours whereas a 2C charge or discharge takes 30 min. Batteries best operate at low C rates, so the lithium ions intercalate smoothly into the electrodes, without damaging them [16]. Additionally, LIBs best perform at a temperature of 25 ± 2 °C [17]. Charging LIBs at low temperatures (below 0 °C) can lead to contraction in the electrode sheets and it can reduce the electronic conductivity of the electrolyte, complicating the Li intercalation into the graphite [16]. Therefore, when batteries have to be charged at low temperatures, it has to be done at a low C rate. In contrast, high operating temperatures (above 100 °C) can cause cell swelling and loss of the battery capacity [13].

Fig. 2 Weight percentages of the LIB components [10]. Copyright 2021 Elsevier

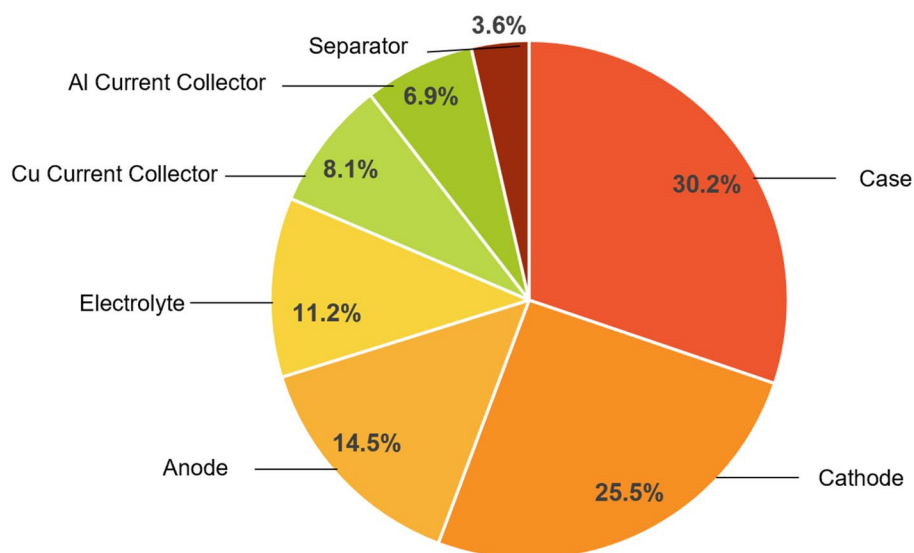
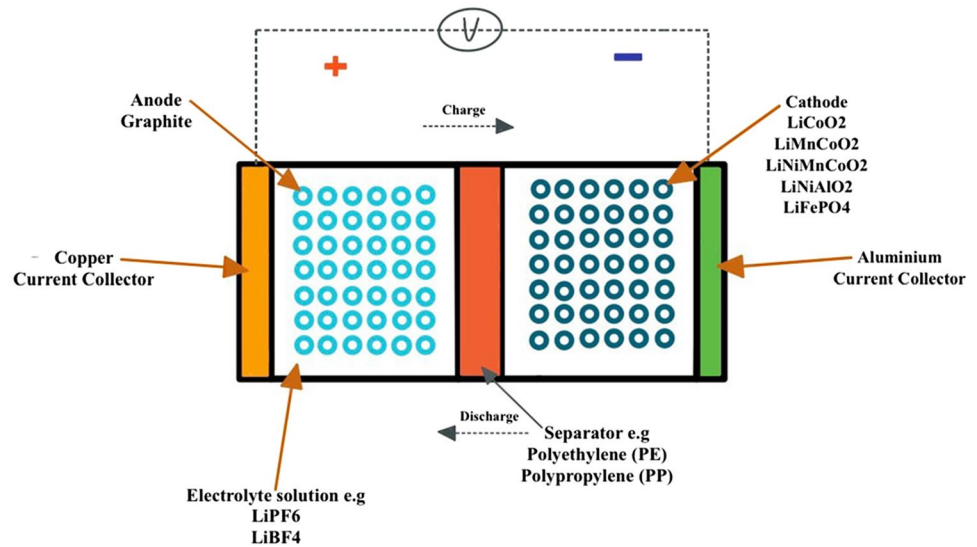


Fig. 3 Schematic representation of the lithium cell battery [15]. Copyright 2021 Elsevier



Characteristics of LIBs

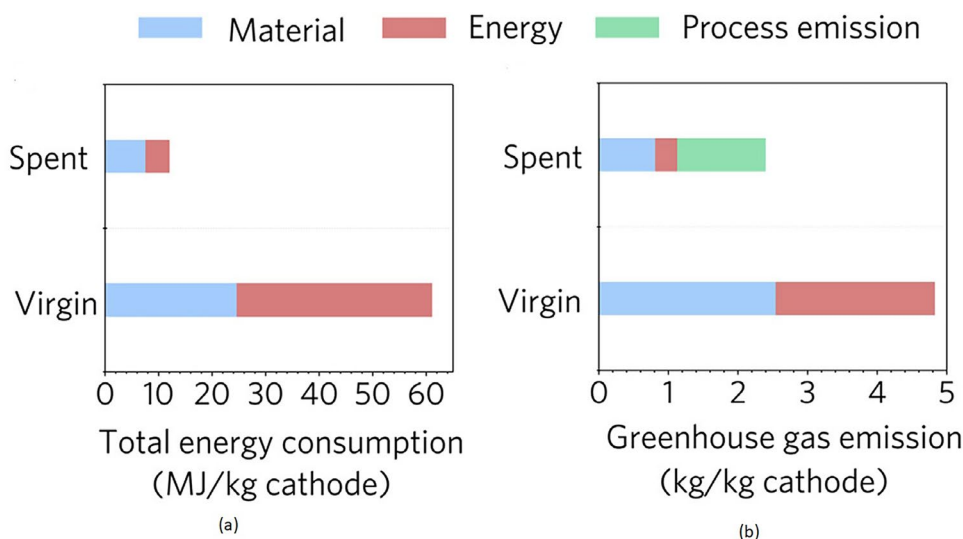
LIBs feature long storage life compared to other types of batteries where they are efficient in charging and discharging; they also have low self-discharge, high energy density (specific energy of 230 Wh/kg) and low weight [18]. However, due to the electrolyte's flammability, LIBs require rigorous testing procedures and protection during production and usage to prevent accident and failure [18]. Depending on the cathode material used, each type of LIB features different characteristics and applications. For example, lithium cobalt oxide (LCO) batteries feature high energy density which makes them suitable in laptops, cameras, and other electronics [19]. On the other hand,

nickel manganese cobalt (NMC) batteries have a long-life cycle and are less expensive than LCO batteries which makes them suitable for e-scooters and some EVs. Lithium manganese oxide (LMO) batteries have higher specific power and thermal stability than LCO batteries, thus, they are used in medical instruments, portable power tools and some EVs like Nissan leaf [20]. Lithium iron phosphate (LFPs) are safe to handle, feature high temperature stability and have a long-life cycle, making them a good option in EVs. However, since LFPs have a relatively low energy density, lithium nickel cobalt aluminum oxide (NCA) batteries are more commonly used in EVs and are known as the battery choice for Tesla [21]. The characteristics of each type of LIB are summarized in Table 1 [22].

Table 1 Characteristics of different types of LIBs [22]

Cathode material	Abbreviation	Nominal voltage (V/cell)	Operating range (V/cell)	Energy density (Wh/kg)	Number of life cycles	Characteristics
Lithium cobalt oxide	LCO	3.60	3.0–4.2	150–200	500–1000	Original commercial type expensive raw materials Relatively high energy density
Lithium nickel cobalt aluminum oxide	NCA	3.70	3.0–4.2	200–260	400–1000	Highest energy density per unit mass
Lithium nickel manganese cobalt oxide	NMC	3.70	3.60–4.0	160–230	2000–3000	Safer and less expensive than LCO Good life cycle Promising technology
Lithium manganese oxide	LMO	3.70	3.0–4.2	100–150	400–750	Safer and less expensive than LCO Poor life cycle
Lithium iron phosphate	LFP	3.20	2.0–3.65	90–160	2000–7000	Very safe High power Lower energy density Best high-temperature stability

Fig. 4 Virgin versus spent batteries' material and energy consumption **a**; virgin vs spent batteries' GHG emissions **b** [30]. Copyright 2020 Elsevier



LIB life degradation

In EV applications, LIBs generally have a lifespan of 5–8 years, varying from manufacturer to another. Additionally, when the batteries' residual capacity is less than 80% of their original capacity, they become dysfunctional in the EVs [8]. LIB degradation restricts the EV's ability to store energy and produce power, as well as its performance. One of the main reasons behind LIB degradation is the side reaction, i.e., Li plating, where Li forms on the anode's surface rather than intercalating into it. This can happen because of fast charging (low C rate), where the high electrolyte potential accelerates the side reaction relative to the main intercalation reaction. Additionally, when the anode's surface becomes totally lithiated, this would limit the available sites for Li, leading to its accumulation on the anode's surface [23], which often occurs at low temperatures [24].

Another reason behind LIB degradation is the accumulation of Solid Electrolyte Interface (SEI) on the anode's surface as a result of contact of the liquid electrolyte with the anode's electron-conductive surface. As the cell ages, the SEI layer keeps accumulating which traps Li ions and prevents them from engaging in charge storage. This is known as loss of lithium inventory. At high temperatures, the SEI accumulation rate increases which leads to the consumption of the electrolyte solvents and thus, the reduction of the electrolyte's conductivity [25]. For graphite anodes, the majority of the SEI production takes place during the first cycle, which typically results in an irreversible capacity loss of 10% or less [26]. Furthermore, the accumulated metallic Li in the SEI could grow and form branch-like structures, called dendrites that can penetrate into the insulating separator and short-circuit the battery [27].

Furthermore, the diffusing Li ions during charging and discharging of the cell induce stress and can deform or

fracture the battery electrodes. This is clearly observed close to the separator due to the higher local current densities, causing larger stresses [23]. Additionally, significant cell degradation can be caused by the corrosion of aluminum current collector. This corrosion is triggered by the electrochemical oxidation of solvent molecules and residual water in the electrolyte [28]. To elaborate, the dissolved aluminum can redeposit on the cathode active material (CAM), preventing Li intercalation and increasing electrical resistance. It can also migrate and deposit on the anode, influencing the performance of the anodic SEI [29].

Feasibility study

To assess the practicality of this recycling process, the total material and energy consumption, and the greenhouse gas (GHG) emissions produced from recycling waste LIBs have to be compared with manufacturing new LIBs from virgin materials. These comparisons are shown in Fig. 4. It can be concluded that making new batteries from virgin materials consumes almost 3 times more material than recycling them. It also consumes around 36 MJ/kg cathode input of energy, which is 9 times more energy than recycling. Additionally, making new batteries from virgin materials produces total GHG emissions of almost 4.8 kg/kg cathode input, where 2.5 are generated due to the materials and 2.3 from energy requirements. On the other hand, recycling produces less total GHG emissions of almost 2.4 kg/kg cathode input, where 1.3 are generated as process emissions, 0.8 from materials and 0.3 from energy [30]. Therefore, recycling waste LIBs is needed to lower the material and energy consumption, and the generation of GHG emissions. This also goes in line with the Net Zero UAE vision, to achieve net-zero emissions in the country by 2050 [31].

Pre-treatment of LIBs

Before recycling, batteries must be pretreated to separate them into their original constituents. Pretreatment involves discharging, dismantling, and separating the cathode materials.

Discharging

Discharging the cell before processing is crucial since the majority of used LIBs that recycling facilities receive are at least partially charged. If one of these LIBs is crushed or opened in air before discharging, severe injury or death could result due to the toxicity of fluorinated gases that may be released. Additionally, discharging the batteries before processing prevents self-ignition and short circuiting [32].

Discharging can be done by different methods. For instance, connecting a used LIB to a discharger can be utilized to discharge the battery to less than 0.1 V. However, this technique does not suit the mass spent LIBs [33]. Another method, which was studied by Torabian et al. [34], utilized different salt solutions such as NaCl, Na₂S, or MgSO₄ as a discharging medium. In this case, the battery's anode and cathode were connected to one end of crocodile clip wires and the other ends were submerged in the solution. Results showed that discharging in NaCl solution is more efficient than Na₂S or MgSO₄. Additionally, submerging a LIB in a 10% NaCl solution leads to a discharge efficiency of 72% in 358 min [34]. Furthermore, discharging the battery in 20% NaCl solution with zinc powder yielded a discharge time of about 20 min. Using Na₂S as a discharging medium increases the rate of corrosion due to the formation of cuprous sulfide [34]. On the other hand, MgSO₄ forms metallic precipitates on the platinum wires which lowers the discharge rate [35]. Nevertheless, the use of NaCl solution causes sedimentation and corrosion of the electrodes as copper corrosion strongly depends on the presence of chloride ions [34].

Another technique is to vertically immerse the tips of the battery inside the salt solution rather than its whole body. This setup minimizes the risk of leakage of the battery minerals into the solution and prevents corrosion. It also reduces the discharge time to less than 5 minutes when using a 5% NaCl solution [34].

Dismantling and mechanical separation

After discharging, dismantling LIBs and manually separating them into cathodes, anodes, organic separators and steel or plastic cases take place. For large scale operations,

separation occurs mechanically, through crushing or sieving or magnetic separation.

In mechanical separation, discharged batteries move through a conveyor belt to a shredder, where the battery structure changes from their original layers into small pieces. These small pieces are composed of metals and plastics. The plastics, which are lightweight, are separated from the metals using air blowers. Then, the metals undergo other separation processes such as magnetic separation and vibrating screen. The process was used by Suny Group in China [36]. Their final products contain mainly copper, aluminum, lithium, cobalt oxide and graphite powder, with powder recovery rate of more than 98% [36]. The recovered powder is also known as black mass in the industry [37].

LIBs main recycling processes

Hydrometallurgical process

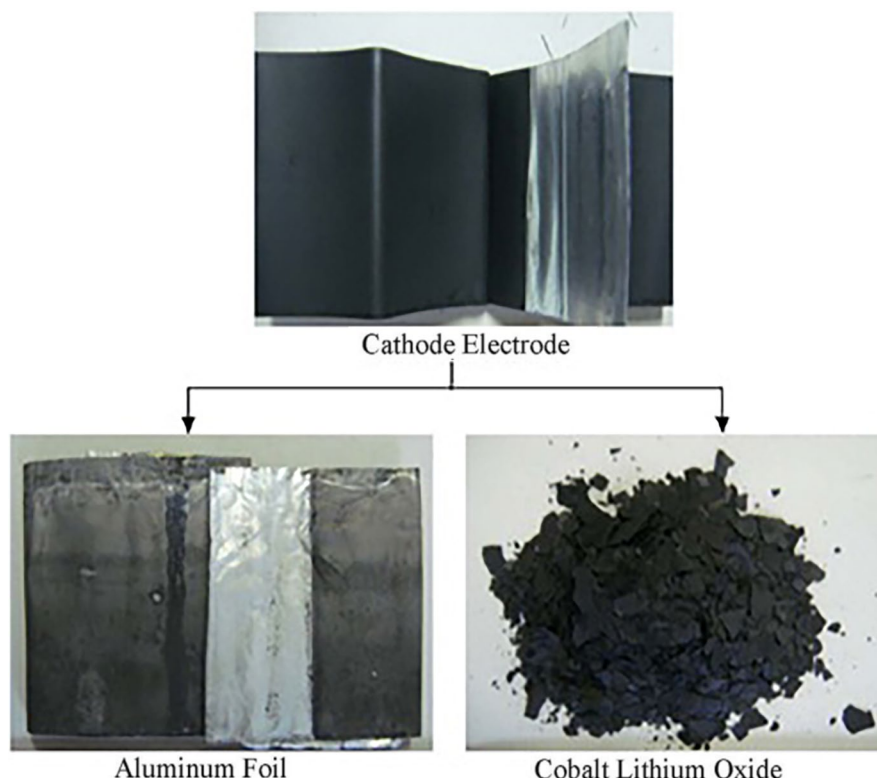
The metal powder that results from pretreatment can undergo a hydrometallurgical process, to further separate the CAM and extract the valuable metals. This can be done through dissolution, or thermal treatment, or vacuum pyrolysis.

Separation of CAM from the aluminum foil

Dissolution In dissolution, the theory of “Like dissolves Like” is used where an organic solvent such as polar N-methylpyrrolidone (NMP) is dissolved into the polar binder, polyvinylidene fluoride PVDF, at 100 °C for 1 h, to separate the CAM from the aluminum foil [38]. Alternatively, the separation can be achieved using 10% NaOH solution at 90°C for 5 h with continuous stirring [39]. In the case of LIBs that use polytetrafluoroethylene (PTFE) binder, trifluoroacetate (TFA) is the most suitable organic solvent, where it is dissolved at a temperature of 40 °C for 3 h [40]. Nevertheless, the dissolution process has some disadvantages. For example, the organic solvents cannot remove all the impurities and the recovered materials are required to undergo a calcining process for further purification. Additionally, the organic solvents are very expensive and do not satisfy a scale-up recycling process [33].

Thermal treatment Thermal treatment is another technique to separate the CAM from aluminum foil. In this case, the cathode is shredded and left in a furnace at elevated temperatures to burn the excess carbon and organic matter. Unlike dissolution, this process is convenient and suitable for large scale recycling processes. However, an excessive amount of toxic gases is emitted during the process and thus additional purification might be required [33]. Furthermore, in the case of LCO batteries, thermal treatment at 900 °C for 1 h can

Fig. 5 Separation of the CAM by vacuum pyrolysis [42]. Copyright 2011 Elsevier



reduce the leaching efficiency, which is the following step in hydrometallurgy [41].

Vacuum pyrolysis Vacuum pyrolysis is an alternative thermal treatment method which weakens the adhesion between the aluminum foil and the CAM by decomposing the organic materials to lower molecular weight products, resulting in the separation of the CAM from the aluminum foil [33]. For instance, an experimental study used a vacuum pump, vacuum furnace, and reactor to perform vacuum pyrolysis on LCO batteries [42]. Initially, the electrodes of the cathodes were placed in a reactor and the pressure of the system was kept below 1 kPa. The reactor was then heated to 600 °C at a rate of 10 °C/min, and the temperature was maintained there for 30 min. The volatiles, leaving the reactor, were condensed and vacuum pumps were used to extract non-condensable gases which were then collected by a gas collector [42]. The results of the separated aluminum foil and CAM (LCO) are shown in Fig. 5.

Leaching

After pretreatment, one way to extract metals from used LIBs is through leaching, which is a major step in hydrometallurgy. Leaching converts the metals in the CAM, collected from the pretreatment process, into ions dissolved in an aqueous solution. It is accomplished by the use of either

inorganic acid, organic acid, or alkali solution as leaching agents.

Inorganic acid leaching Joulie et al. [43] investigated leaching using different inorganic acids (H_2SO_4 , HCl and HNO_3) by placing the cathode material into a 100 mL one neck glass reactor, immersed in an oil bath for temperature control. They concluded that the most efficient inorganic acid for leaching is HCl [43]. However, chloride ions may form due to HCl oxidation which cause threats to the environment and may need further treatment, increasing the cost of the recycling process. Sulfuric acid and nitric acid, H_2SO_4 and HNO_3 , are used in the presence of a reducing agent such as hydrogen peroxide (H_2O_2). According to several studies, the addition of H_2O_2 enhances the leaching efficiency as it converts insoluble cobalt (III) materials to soluble cobalt (II) [33]. Additionally, H_2SO_4 may be utilized with vitamin C which helps in reducing Co (III) to Co (II), thus, enhancing the leaching efficiency [44]. Similarly, H_2SO_4 with SO_2 at 85 °C, 3 mol/L H_2SO_4 concentration for 1 h, improves the leaching efficiency to 99.4% [45]. For optimal leaching rate, the leaching acid's concentration, the solution's temperature, the solid to liquid ratio (S/L), the time of the reaction, and the addition of a reducing agent must be determined [46]. Table 2 summarizes the leaching efficiency of various inorganic acids for different types of LIBs at specific conditions.

Table 2 Summary of different inorganic acid leaching agents and their leaching rates

Type of LIBs	Leaching agent	Temperature (°C)	S/L ratio (g/L)	Time (h)	Leaching Efficiency (%)	Refs
LiCoO ₂	2 M H ₂ SO ₄ + 2.0 vol % H ₂ O ₂	60	33	2	Co: 96.3; Li: 87.5	[47]
LiCoO ₂	2 M H ₂ SO ₄ + 5.0 vol % H ₂ O ₂	80	50	1	Co: > 99; Li: > 99	[42]
LiCoO ₂	2 M H ₂ SO ₄ + 5.0 vol % H ₂ O ₂	75	100	1	Li: 99.1; Co: 70.0	[48]
LiCoO ₂	2 M H ₂ SO ₄ + 8 vol % H ₂ O ₂	75	50	1	Co: 98	[49]
LiCoO ₂	2 M H ₂ SO ₄ + 6 vol % H ₂ O ₂	60	100	1	Co: 98; Li: 97	[50]
LiCoO ₂	3 M H ₂ SO ₄ + 0.4 g/g cellulose	95	25	2	Co: 54; Li: 100	[51]
LiCoO ₂	3 M H ₂ SO ₄ + 0.4 g/g sucrose	95	25	2	Co: 96; Li: 100	[51]
LiCoO ₂	3 M H ₂ SO ₄ + 0.4 g/g glucose	95	25	2	Co: 98; Li: 96	[51]
LiCoO ₂	4.0 M HCl	80	–	2	Co: 99; Li: 97	[52]
LiCoO ₂	1.0 M HNO ₃ + 1.7 vol % H ₂ O ₂	75	10	0.5	Co: ~ 99; Li: ~ 99	[53]
LiCoO ₂	0.7 M H ₃ PO ₄ + 4 vol % H ₂ O ₂	40	50	1	Co: 99.7; Li: 99.9	[54]
LiFePO ₄	2.5 M H ₂ SO ₄	60	100	4	Li: 97; Fe: 98	[55]
Li ₄ Ti ₅ O ₁₂	4 M H ₂ SO ₄ + 20 vol % H ₂ O ₂	80	25	4	Ti: 98; Li: 97	[56]
Mixture	1 M H ₂ SO ₄	95	50	4	Co: 66.2; Li: 93.4 Ni: 96.3; Mn: 50.2	[57]
Mixture	1 M H ₂ SO ₄	95	50	4	Li: 93.4; Co: 66.2 Ni: 96.3; Mn: 50.2	[58]
Mixture	1 M H ₂ SO ₄ + 5 wt % H ₂ O ₂	95	50	4	Li: 94.5; Co: 79.2 Ni: 96.4; Mn: 84.6	[58]
Mixture	2 M H ₂ SO ₄ + 6 vol % H ₂ O ₂	60	100	1	Co: 99	[59]
Mixture	4 M H ₂ SO ₄ + 10 vol % H ₂ O ₂	85	100	2	Co: 95; Li: 96	[60]
Mixture	1 M H ₂ SO ₄ + 0.78 wt % NaHSO ₃	95	20	2	Li: 96.7; Co: 91.6 Ni: 96.4; Mn: 87.9	[58]
Mixture	4.0 M HCl	80	20	1	Co: 99.5; Li: 99.9 Ni: 99.8; Mn: 99.8	[61]

Organic acid leaching Although inorganic acids demonstrate a higher leaching efficiency where more than 99% of Co and Li can be recovered, they release hazardous gases such as Cl₂, SO₃, and NO_x. Therefore, researchers were motivated to investigate organic acids such as citric acid and oxalic acid due to their ease of degradation, recycling, and seldom environmental risks [33]. Additionally, citric acid yields high leaching efficiency where almost 100% of lithium can be recovered as shown in Table 3. Moreover, tannic acid proved to be an effective leaching reagent by promoting cobalt solubility for LCO batteries [62]. Additionally, acetic

acid can be used along with tannic acid as a pH regulator [62]. Tartaric acid can also be used as a leaching reagent in LCO batteries along with H₂O₂, where it acts as a precursor to form cobalt oxalate [63]. However, it has been shown that the use of organic acids for leaching may not be appropriate as they are more expensive than inorganic acids, have slow leaching speed, and have a S/L ratio lower than the inorganic acids which results in a low treatment capacity of organic acids [33]. This prohibits the recycling process of used LIBs on a large scale.

Table 3 Summary of different organic acid leaching agents and their efficiencies [8]

Leaching agent	Leaching condition	Reduction agent	Leaching efficiency
1.5 M malic acid	40 min + 363 K + 20 g/L	H ₂ O ₂	~ 100% Li, 90% Co
1 M oxalic acid	150 min + 368 K + 15 g/L	N/A	98% Li, 97% Co
1.25 M citric acid	30 min + 363 K + 20 g/L	H ₂ O ₂	~ 100% Li, > 90% Co
1.5 M succinic acid	40 min + 343 K + 15 g/L	H ₂ O ₂	96% Li, 100% Co
3.0 M trichloroacetic acid	30 min + 337 K + 50 g/L	H ₂ O ₂	99.7% Li, 91.8% Co, 93.0% Ni, 89.8% Mn

Alkali leaching Alkali leaching can be done using ammonium chloride and sulfuric acid. The use of sulfuric acid increases the leaching efficiencies of Co, Li, Mn and Ni from 0 to 2.5 mol/L. According to an experimental study, leaching efficiency increases with an increase in the concentration of H_2SO_4 or NH_4Cl [64], with optimum H_2SO_4 and NH_4Cl concentrations of 2.5 and 0.8 mol/L, respectively [64]. The experiment was done at a temperature of 353 K with agitation speed of 400 rpm and S/L mass ratio of 100 g/L for 60 min. This type of leaching recovered 99.11% of Li, 97.49% of Ni, 97.55% of Co and 97.34% of Mn [64]. Furthermore, a ternary mixture of 1.5 M ammonia, 1 M ammonium sulfite and 1 M ammonium bicarbonate can be used for leaching, where ammonium bicarbonate can act as a pH buffer to lower the pH variation during the reaction. The reaction was performed at a temperature of 60 °C and S/L mass ratio of 20 g/L for 180 min [65]. However, this type of leaching resulted in lower recoveries of Li and Co, namely 60.53% and 80.99%, respectively [65]. Other alkali leaching agents were studied and the results are summarized in Table 4 [8].

Separation of valuable metals

Once the cathode active materials are leached, the leachate will contain valuable metals such as Li, Co, Ni, Mn, Cu, Al, and Fe. Such methods as solvent extraction, chemical precipitation, and electrochemical deposition can be used to extract the valuable metals from the solution. Two or more methods must be used simultaneously due to the complexity of the solution.

Solvent extraction Due to the high selectivity of solvents for various metal ions, solvent extraction has been utilized for most hydrometallurgy processes. Several solvents such as di(2-ethylhexyl) phosphoric acid (D2EHPA), Mextral 272P, bis(2,4,4-trimethyl-pentyl) phosphinic acid (Cyanex 272), P507, etc., have been used to extract the valuable metals from the solution. For instance, Cyanex 272 is an excellent solvent in extracting Co with a recovery rate of 95–98% [33]. Moreover, using Cyanex 272 with 5 vol% tributyl phosphate (TBP) and kerosene leads to 85.42% of Co extraction at pH 5. On the other hand, using Cyanex 272 with isodecanol yields 99.9% Co extraction at pH 5 [66].

Furthermore, 0.4–0.5 M D2EHPA diluted in kerosene can be utilized to recover 97% Mn at pH of 2.2–5 [67]. To further enhance the selectivity of metals in the extraction process, two or more solvents are mixed and used. This can be referred to as synergistic extraction.

Another common extractant is TBP diluted in kerosene. Kerosene is utilized to reduce TBP's viscosity and density, hence enhancing mass transfer. Additionally, FeCl_3 is used as a co-extractant in the process to enhance extraction [68]. It is also reported that Li recovery rate can reach above 99.9% when three-stage counter-current extraction is used [69].

Chemical precipitation Chemical precipitation is a process in which compounds containing special anions, such as OH^- , $\text{C}_2\text{O}_4^{2-}$, and CO_3^{2-} , form precipitates when added to a leachate solution. This method can be used to separate the valuable metals from the leachate solution. This is applicable because of the solubility differences of the precipitates. Due to the complex structures of the spent LIBs, the recovery of metals is possible by various precipitation reactions that can be controlled by the manipulation of the pH of the solution [46]. Studies showed that the precipitation at a pH 5.5 leads to Li_2CO_3 recovery rate of more than 98% [67]. Moreover, Wang et al. [39] studied the separation and recovery of valuable metals such as Ni, Mn, Co, and Li from used LIBs. They concluded that Mn was recovered as MnO_2 and $\text{Mn}(\text{OH})_2$ using a KMnO_4 reagent, and $\text{Co}(\text{OH})_2$ was selectively precipitated by adding 1 M NaOH solution to achieve an alkaline pH of 11. Then, saturated Na_2CO_3 was added to precipitate the residual Li_2CO_3 aqueous solution. Their results also indicated that Co, Li, Ni, and Mn were all recovered to varying degrees, namely 96.294 wt%, 96.97 wt%, 97.43 wt%, and 98.23 wt%, respectively. To reduce the precipitation steps, coprecipitation is often utilized to precipitate two or more metals simultaneously. This technique is often linked with solvent extraction [39].

Electrochemical deposition Electrochemical deposition separates metals from a solution through differences in electrode potential. Several studies considering this method are available in literature; however, they mostly focus on the recovery of cobalt [33].

Table 4 Summary of different alkali leaching agents and their efficiencies [8]

Leaching agent	Leaching condition	Reduction agent	Leaching efficiency
4 M NH_3 + 1.5 M $(\text{NH}_4)_2\text{SO}_4$	300 min + 353 K + 10 g/L	Na_2SO_3	95.3% Li, 80.7% Co, 89.8% Ni, 4.3% Mn
3 M $(\text{NH}_4)_2\text{SO}_4$	120 min + 453 K + 83 g/L	$(\text{NH}_4)_2\text{SO}_3$	98% Li, 81% Co, 98% Ni, 92% Mn

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Table 5 Summary of the chemical processes for LIP recycling

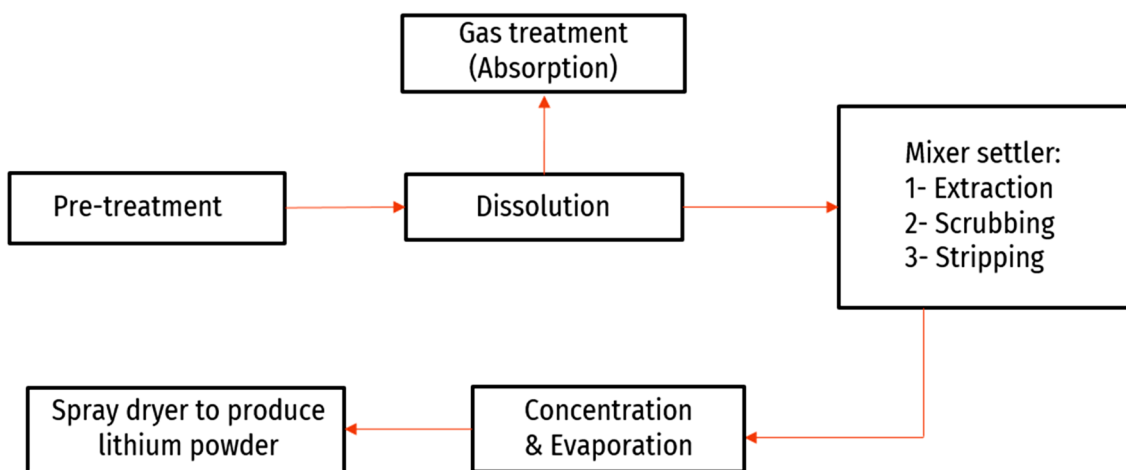
Chemical Processes	Methods/Reagents	Features	Results
Solvent Extraction	Mextral272P Cyanex 272 D2EHPA	Equilibrium pH of 4.5; 20 vol% Mex-tral272P; 300 s; A:O = 1.1 50% saponified 0.4 M Cyanex 272; pH 5.5–6.0, A:O ratio = 1/2 70–75% saponification rate; 20 vol% D2EHPA; pH 5, A:O ratio 0.5	Co: 97.8% [70] 95–98% Co extraction ~ 1% Ni extraction [50] Mn: 97% [33]
Chemical Precipitation	(NH ₄) ₂ C ₂ O ₄ and Na ₂ CO ₃ Na ₂ CO ₃ Multi-step precipitation process	Initial pH 2; 50 °C; 60 min pH adjusted to 2 0.2 M dimethylglyoxime (DMG) solution, 0.5 M oxalic acid and 0.5 M phosphoric acid; 0.5 M KMnO ₄	94.7% of Co and 71.0% Li [47] Fe and Li [59] 99%, 91%, 92% and 94% for Li, Ni, Co and Mn [71]
Electrochemical Deposition	The initial charge and discharge capacity = 130.8 and 127.2 mAh g ⁻¹ Deposited Co ²⁺ as Co(OH) ₂ on a titanium electrode plate; constant potential; 200 °C		First cycle charge efficiency of 97.2% After 30 cycles, charge efficiency of 99.1% [72] Co ₃ O ₄ [73]

Table 5 summarizes some chemical processes in terms of reagents used, features, and results to separate the valuable metals from the cathode.

Figure 6 summarizes the hydrometallurgical process. The LIBs first undergo pretreatment, where they are discharged and dismantled. Then, dissolution takes place to separate the CAM, and any gas produced can be fed to an absorption unit for gas treatment. The solution is then pumped to a mixer settler which is mainly composed of three stages: extraction, scrubbing, and stripping [74]. In extraction, only lithium is extracted from the aqueous phase to the organic phase by the use of extractants such as D2EHPA, Cyanex272, or TBP–FeCl₃ diluted in kerosene. The organic phase becomes rich with lithium whereas the aqueous phase will be almost free of lithium. Next, scrubbing occurs to wash the aqueous

contaminant out of the organic phase. Lastly, lithium is stripped from the organic phase to the aqueous phase [74]. Then, the leftover water is evaporated, and a slurry forms. To convert the slurry into lithium powder, a spray dryer is used where the slurry is in countercurrent flow with hot air.

A study proposed by Wesselborg et al. [68] investigates the use of TBP–FeCl₃ diluted in kerosene to directly extract Li from LIB waste leachate. Two countercurrent stages were used to result in a Li purity of 65.6% in the loaded organic phase. To further enhance the Li purity, a single stage scrubbing takes place with 1 M LiCl + 2 M AlCl₃ which yields Li purity of 98.9%. Then, three-stage stripping occurs with 6 M HCl as a stripping agent to achieve Li purity of 99.1%. The stripped organic phase can then be recycled after saponification with NaOH or NaCl.

**Fig. 6** Block flow diagram of the hydrometallurgical process

Commercial LIBs recycling plants

Fortum, a Finnish energy company, is constructing a hydrometallurgical plant in Harjavalta, in Finland, to recycle spent LIBs. The plant is expected to be ready by the beginning of 2023 and will focus on increasing the capacity of recycling battery materials [75]. The company uses both mechanical and hydrometallurgical approaches with low carbon emissions to recover Li, Ni, Co, and Mn from the spent batteries, which can be then reused to manufacture new batteries. During the mechanical process, the batteries are first discharged and disassembled at Fortum's plant in Ikaalinen, Finland. Plastics and black mass are retrieved. The black mass is then collected to undergo hydrometallurgy in Fortum's plant in Harjavalta. Fortum reported that 80% of the battery can be recycled through these processes and 95% of the black mass's valuable metals can be recovered via hydrometallurgy. Another company that uses the same process is Primobius located in Germany [76].

Table 6 summarizes the advantages and disadvantages of the hydrometallurgical approach. Some of the advantages include application and separation process flexibility [77]. Furthermore, hydrometallurgy involves low energy consumption, high metal recovery rate, and high product purity [42]. However, some of its drawbacks include the consumption of high chemical reagents and the long duration of the process [8]. In addition, it is uneconomical for LFP batteries, and it also generates a high volume of wastewater that needs to be further recycled or disposed [77].

Pyrometallurgical process

Metals can be extracted and refined via pyrometallurgical process. Incineration, calcination, and pyrolysis are the thermal pretreatment techniques for recovering the CAM, and roasting or smelting processes are used to process the enhanced metal fraction. Pyrometallurgical recycling systems have been widely used in several industries, such as the ultra-high-temperature smelting technology (UHT) of Umicore in Belgium, Glencore (Xstrataroasting)'s and

smelting process, Switzerland Inmetco's high-temperature melting recovery (HTMR) technique, Germany's Accurec, and Sony-calcination Sumitomo's method [15].

Thermal pretreatment techniques

Utilizing heat pretreatment techniques to disintegrate the battery module and separate it into enriched metal fractions that may be recovered by extractive metallurgy are current trends in recycling used LIBs. These techniques are described below.

Incineration pretreatment During the incineration process, the carbon and the binder are burned in the presence of air or oxygen at high temperatures. In this case, all organic matter and plastic covers are burned. The ideal temperature range for incineration can affect the recovery rates of Co and Li. For example, incineration of LiCoO_2 at 700 °C for 60 min improved the Co and Li recovery rate due to the carbothermic reduction (CTR) and carbon elimination. After removal of most of the carbon, the rate at which Li was recovered during the leaching process increased because carbon would act as an adsorbent for Li salts [15].

Additionally, the organic matter and carbon compounds in the used LIBs are hydrophobic and, thus, cause issues with the subsequent solid–liquid separation steps. However, increasing the incineration temperature further causes the aluminum foils to melt, covering the LiCoO_2 particles which limits the recovery of Co. Hanisch et al. [15] developed the Adhesion Neutralization via Incineration and Impact Liberation (ANVIL) method to improve the separation of the current collectors from the active cathode material and addressed the issues caused by the melting of the Al foil. In this process, the PVDF is broken down using a moderate temperature of 500 °C, then the coating powder and current collector foils are separated using an air-jet separator to avoid any interference with the process.

Pyrolysis pretreatment Pyrolysis is the process of thermally converting organic compounds into low-molecular weight

Table 6 Advantages and disadvantages of hydrometallurgy process for LIB recycling [77]

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

products that can be utilized as fuel or chemical feedstock. This is normally done by heating the LIB material above its breakdown temperature in an oxygen-free or inert atmosphere to prevent occurrence of any unfavorable chemical reactions as lithium is highly reactive with air. It is also used to eliminate any remaining electrical energy in the battery. Additionally, during pyrolysis, the electrolyte is lost but can be recovered by condensation. On the other hand, the CAM can withstand the pyrolysis temperature and remain as solid residue which can be treated further during the following recycling procedures. The volatile gases produced from pyrolysis are fed to a condenser and collected, whereas the non-condensable gases are extracted using a gas collection system using a vacuum pump. Additionally, the aluminum foil gets crisp under pyrolysis which simplifies its detachment from the cathode [15].

Pyrolysis can be carried out in a packed bed reactor where such metals as Ni, Co, Mn and Li can be recovered from waste LIBs. The input material (black mass) is fed from the top onto the hot graphite bed. The bed is heated inductively by the surrounding copper coils. To elaborate, the induction coil at the top is used to melt the black mass and form a thin molten layer as it moves downward. The second induction coil, placed halfway of the reactor is used to ensure that the reduction reaction takes place. The last induction coil at the bottom is used to maintain a reaction temperature above the melting temperature of the mixture [78]. The gaseous products move through the flue gas pipe to be further treated, whereas the liquid product flows downward. It is worth mentioning that this reactor offers a large surface area for the reaction to take place, and good temperature control over different zones of the reactor [78]. This type of reactor reveals 95% Li recovery and over 64% of phosphorus when using LFP batteries [79].

Extractive pyrometallurgical process for recycling LIBs

Roasting/calcination After pretreatment, the recovered CAM is then processed by CTR roasting, which involves exothermic reactions between gas and solid at high temperatures. In CTR, the CAM is heated in presence of a reducing agent such as carbon, charcoal, or coke, leaving carbon residue and a combination of alloys/intermediate compounds (impure metals/oxides) for further refinement. It is reported that CTR is able to destruct oxygen octahedrons in the crystal structures of lithiated metal oxide, which facilitates its reduction [80]. Once the oxygen octahedrons get destructed, Li and Co effortlessly escape, intensifying the reaction. Furthermore, when carbon is utilized as a reducing agent, metals can be reduced in a double redox reaction at relatively low temperatures. It is worth mentioning that the presence of carbon in CTR does not diminish the CAM directly; instead, a series of coupling processes take place [80].

Smelting Smelting is another efficient pyrometallurgical method used for extraction of precious metals from spent LIBs. During this process, the battery material is heated above its melting point to enable the reduction-mediated separation of the metals in the liquid phase and the subsequent production of immiscible molten layers. In this process, the used batteries are fed into a furnace at a high temperature without being pretreated [15]. Smelting involves two steps: (i) heating the material at a low temperature to evaporate the electrolyte and prevent burst (since intense heating could cause the battery to explode due to overpressure caused by the electrolyte's sudden evaporation), and (ii) heating the material at a higher temperature to melt off the plastic as part of the battery. After smelting, the transition metals (Co, Ni, and Cu) are preferentially condensed into a molten alloy phase, sinking to the bottom of the furnace, and then fed to the molten metal pool. However, lithium oxide present in the cathode does not get reduced and is reported as a leftover byproduct, which is referred as slag [15]. To further recover Li from the slag, hydrometallurgical techniques are used. Since Li and Mn are lost in the slag during the smelting process, batteries with high Co and Ni content are considered more cost-effective than current Mn spinel oxides or LFP batteries. From an industrial-scale point of view, reduction smelting is frequently used due to its straightforward operation and high productivity [15].

The pyrometallurgical process to recover lithium from spent LIBs was presented by Bae and Kim [81]. In this process, the CAM passes through calcination to obtain lithium carbonate and lithium metal oxide, where the metal can be Co, Mn or Ni. After calcination, the heat-treated product passes through water leaching, then filtration to obtain the lithium carbonate solution as filtrate, and the metal oxides as solid residue which can be separated and further treated to recover the metals. Then, the water solution that has lithium carbonate passes through water evaporation to obtain the lithium carbonate powder that can be used in battery re-synthesis [81].

Oxygen-free roasting and wet magnetic separation

Another process that can be classified under pyrometallurgy for recycling spent LIBs is the oxygen-free roasting and wet magnetic separation. In this process, the spent LIBs undergo discharging, scraping, screening, oxygen-free roasting, and finally wet magnetic separation.

The LIBs are first discharged in a 5 wt% NaCl solution for one day. Then, it is dried by air. After that, the discharged LIBs have to be crushed using a hammer mill and electric screener. The powder is then fed to the Carbolite single tube furnace (STF), where oxygen-free roasting occurs. The system includes a heating unit and the Carbolite STF, which can achieve a maximum operating temperature of 1500 °C. The

left flange of the tube connects the inlet valve with a nitrogen vessel to fill the tube with nitrogen, whereas the right flange connects the outlet valve to the vacuum pump. Inside the flanges, condensed water circulates to protect the O-ring seal from melting at high temperatures [82].

After roasting, a magnetic stirrer is used to separate the products from the residue, where the ferromagnetic material (Co) in the powder gets attached to the magnetic stirrer as it is fed, while the graphite gets collected as a deposit by filtration. Then, the remaining solution is evaporated to collect Li_2CO_3 . The recovery rate of Co, Li, and graphite were 95.72, 98.93, and 91.05%, respectively [40].

Table 7 summarizes the advantages and disadvantages of pyrometallurgical recycling [77]. Some of the advantages include high rate of chemical reactions, large treatment capacity, flexibility in the feed material, ease of operation, and less environmental impact from the dross [15]. On the other hand, pyrometallurgical recycling is not applicable on LFP batteries and it requires high energy and capacity [77].

Direct recycling of LIB cathodes by targeted healing

Direct recycling is another technique that is used to recycle waste LFPs. LFP is a type of LIB that uses LiFePO_4 as a cathode and graphitic carbon as an anode. Although these batteries provide low energy density, they are less expensive and provide safe and fast charging, which makes them a popular choice for EVs. For example, Tesla confirmed that nearly half of its EVs are relying on LFP batteries [83]. Additionally, Elon Musk has stated that Tesla plans to shift more EVs to LFP batteries to overcome the Ni and Co supply problems [83]. Due to its growing demand, recycling used LFPs safely and economically has become a concern. One of the reasons that leads to a drop in the performance of the LFP battery is the switch of Fe and Li ion positions in the crystal structure. As a result, when the Li ions are released from the anode to the cathode during battery discharging, their pathway may get blocked [84]. A study led by Xu et al. [30] suggested a targeted healing process to force the Fe atoms back into their place, and free the pathway for Li ions. To investigate this scenario,

researchers studied commercial batteries that had lost half of their capacity. Those batteries were disassembled, and the cathode powder was harvested. Then, the relithiation step took place, where the cathode powder was placed in a 100 mL reactor filled with 80 mL of 0.2 M LiOH and 0.08 M of citric acid solution [30]. The powder was placed in the solution for 1–5 h at a temperature of 80 °C. In this step, citric acid acts as a reducing agent that transfers electrons to the Fe ions, which reduces the electronic repulsion forces, allowing Fe ions to go back to their original spots. After relithiation, the powder has to be washed with deionized water and dried. The dry powder is then mixed with excess 4% of Li_2CO_3 and thermally annealed [30]. Annealing is done at a temperature of around 600 °C for 2 h in nitrogen, with a temperature ramping rate of 5 °C/min. After annealing, the powder has to be cooled back. This step of heating and cooling improves order and stability of the cathode crystalline structure [85]. Finally, the regenerated cathode material can be used directly by cell manufacturers without further re-synthesis, unlike the pyrometallurgical and hydrometallurgical methods where large quantities of cathode material is completely lost [30]. On the other hand, direct recycling is a non-destructive process that maintains the cathode value.

Another advantage of this method is that the relithiation solution can be recycled. Some researchers attempted a second batch of LFP batteries relithiation using the same used LiOH and citric acid solution [30]. Their results showed that the capacity and stability of the second batch LFP batteries is at the same level as the first batch [30]. Furthermore, direct recycling helps in reducing GHG emissions by 11–28%. The advantages and disadvantages of this process are summarized in Table 8 [77]. Although this process seems to have many advantages, it is still under investigation and has not been scaled up to a commercial recycling plant. It also focuses on one type of LIB, but researchers are optimistic that it can be applied to LCO batteries as well [86]. In LCO batteries, relithiation can occur using Li_2CO_3 solution. In this step, Li_xCoO_2 decomposes to Co_3O_4 which reacts with Li_2CO_3 to form LiCoO_2 back, as shown in the equations below [87].

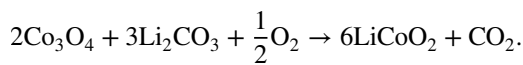
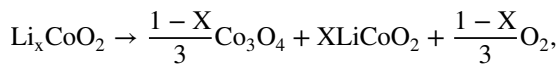
Table 7 Advantages and disadvantages of pyrometallurgy process for LIB recycling [77]

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

Table 8 Advantages and disadvantages of direct recycling [77]

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

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Precise separation process

An effective technique to recycle LIB cells without pretreatment is known as precise separation. Figure 7 describes the precise separation of LIBs which involves immersion in water, where the separated materials are not mixed and can be easily extracted from the aqueous solution. When submerged, the only material in the LIB cell that reacts with water is lithiated graphite (LiC_x) that present in the anode, enabling the anode materials to separate from the copper foil, and produce soluble LiOH , which makes it easy to recover Li [32]. A group of researchers [32], in Umea University in Sweden, performed precise separation by disassembling the LIB cell in water as portrayed in Fig. 8.

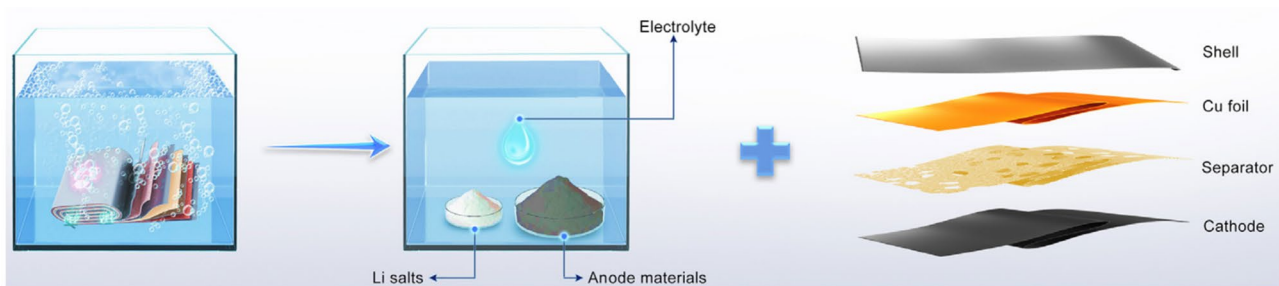
In this process, the cell shell was first removed, and the jellyroll was opened (Fig. 8a). The separated cathode/separator and copper foil are shown in Fig. 8b. Figure 8c displays the anode materials that have been removed from the aqueous solution. The aqueous solution is shown in Fig. 8d. After the anode materials have been removed, Na_3PO_4 was

added, and a filter has been used to remove the precipitated Li_3PO_4 . The LIB cell can, thus, be separated into five distinct solid parts, namely, shell, separator, cathode, copper foil, and residual anode materials, as well as an aqueous solution containing Li , electrolyte, and water-soluble binder. The separator material can subsequently be used to further isolate the cathode. Only a small quantity of Li , from the SEI components, was present in the separated graphite; most of the Li resulted from the anode and electrolyte in the aqueous solution. The electrolyte dispersed in a vast volume of water can be converted into new compounds, preventing potential Li loss and fluorine emission [32].

The clear solution left behind from Li precipitation can be utilized to disassemble another cell without wasting resources or producing waste [32]. However, this process imposes a separation challenge due to the structural complexity of LIB cells and the safety risks associated with their constituent materials when they are partially charged. Such cells should be deconstructed in a controlled manner in an environment that reduces the dangers associated with this reactivity since they will have a high content of highly reactive LiC_x . Furthermore, the cathode, separator, and anode are packed closely together in a single assembly. Therefore, another non-destructive technique for peeling the cathode or the anode from this assembly is required. Lastly, the poisonous, flammable, and volatile electrolyte, which contains nonvolatile Li salts completely wets all cell components, making precise separation more difficult [32].

Other process for recycling waste libs: eddy current separation for recovering aluminum and LFP component

Another technique to recycle LFPs is via eddy current separation. In this process, the spent LFPs are first discharged then manually dismantled into aluminum shell, lead, and inner core [88]. The aluminum shell is directly recycled, whereas the inner core is separated into anode slices, cathode slices and diaphragm. The diaphragm is disposed of by a downstream company. The anode slices are scraped to

**Fig. 7** Schematic of precise separation process [32]. Copyright 2022 Elsevier

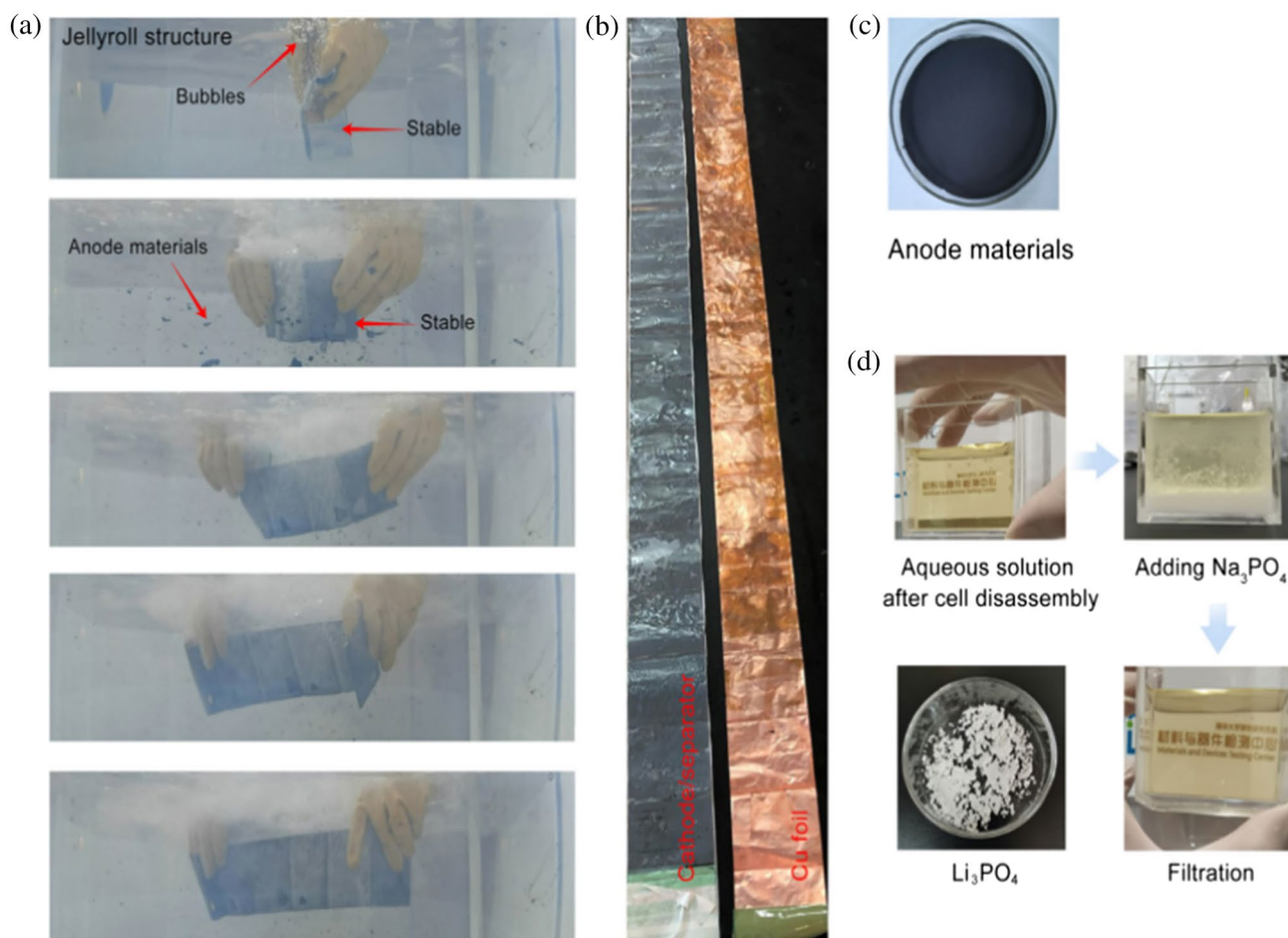


Fig. 8 Disassembly of LIB in water via precise separation [32]. Copyright 2011 Elsevier

separate the copper foil from the anode powder. The cathode gets thermally treated to weaken the adhesion forces between the cathode powder and the aluminum foil. Thermal treatment was done at 240 °C and then cooled to room temperature for 120 min. After that, the cathode is shredded using a shear type shredder. The shredded cathode is then fed to an eddy current separator (ECS) to separate nonferrous metals (aluminum) from the LFP mixture. ECS mainly depends on the magnetic field's ability to levitate the conducting component. Therefore, the ratio of conductivity to density indicates the degree of separation by eddy current. For instance, aluminum has a high ratio of conductivity to density which enables its separation by eddy current from the LFP mixture [89]. The feeding speed to the ECS is 1 m/s and the magnetic field rotation speed is 4 m/s. This method showed a 92.52% recovery rate of the LFP mixture.

The design of a typical commercial ECS is described by Smith et al. [90]. A cylindrical array of permanent magnets with alternating polarities is rotated to create a time-varying magnetic field. When the magnetic field is strong and fast enough, the nonferrous metals (aluminum) and nonmetals

(LFP) leave the feed belt and are separated by a mechanical splitter [90].

Comparisons between hydro, pyro, direct recycling, and precise separation

Hydro, pyro, direct, and precise separation processes can be compared in terms of energy and water consumption, gas and solid waste generated, and total recycling efficiencies. Accordingly, the energy consumption of hydro is around 5 MJ/kg cell input, which is less than pyro, precise separation, and direct recycling (13, 6, and 43 MJ/kg cell input, respectively) [32]. Direct recycling consumes 3.9 L of water, which is comparable to the other methods (pyro: 2.6 L, hydro: 9.5 L, and direct: 14.0 L). More importantly, the precise separation produces only 0.28 kg of waste gas per kg cell input, whereas pyro, hydro, and direct methods produce more waste gas (2.0, 1.5, and 1.8 per kg cell input). The total recycling efficiency of precise separation is 97.82% which is higher than that of pyro, hydro and direct methods (12.32, 37.54, and 64.5%, respectively) [32].

Figure 9 shows the process steps for each method to produce cathodes. Spent batteries can undergo either pyro, hydro, or direct recycling. In pyro, smelting occurs first, then precipitation to form precursors which are then dried and annealed to obtain the cathode powder. On the other hand, hydro and direct require physical separation before processing [30].

Table 9 summarizes the advantages, disadvantages, and challenges for hydro, pyro, and direct recycling processes [91].

Proposed recycling process

Drawing from the preceding conversation, this analysis presents a hydrometallurgical process as the most appropriate scheme for recycling scheme for spent lithium batteries. The hydrometallurgical process consumes less energy as it operates at lower temperatures and requires fewer raw materials compared to pyrometallurgical processes

[87]. While pyrometallurgical and direct methods generate excessive solid and gas waste due to smelting and annealing, the hydrometallurgical process produces less waste [88]. The pyrometallurgical process is the least environmentally friendly process due to the emissions from smelting, while the hydrometallurgical process, for instance, can achieve high recovery rates of valuable materials with minimal emissions and energy consumption [92]. The direct recycling process, on the other hand, involves targeted healing of LIB cathodes, reducing the need for new materials and minimizing the environmental impact [93]. In addition, the hydrometallurgical process is known for its ability to extract valuable metals with high purity levels, making it highly desirable for recycling waste LIBs [94].

The proposed hydrometallurgical process is shown in Fig. 10. To commence, the batteries are fully discharged through immersion in a sodium chloride solution. Subsequently, the batteries are conveyed to a shredder, where their structure is transformed from original layers into smaller fragments comprising metals and plastics. Employing air

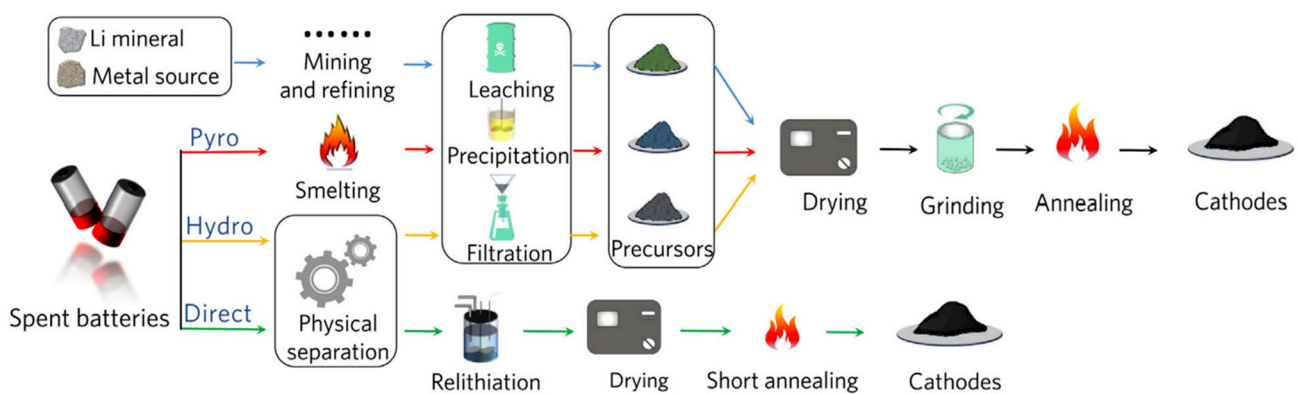


Fig. 9 Processes for pyro, hydro, and direct LIB recycling processes [30]. Copyright 2020 Elsevier

Table 9 Advantages, disadvantages and challenges of hydro, pyro and direct recycling processes [91]

Process	Advantages	Disadvantages	Challenge
Hydro	High recovery rate High purity product Low energy consumption Less waste gas High selectivity	More wastewater Long process	Wastewater treatment Optimize the process
Pyro	Simple operation and short flow No requirement for categories and the size of inputs High efficiency	Li and Mn are not recovered High energy consumption Low recovery efficiency more waste gas and the cost of waste gas treatment	Reduce energy consumption and pollution emissions Reduce environmental hazards Combine hydrometallurgy well
Direct	Short recovery route Low material consumption Environmentally friendly High recovery rate	High operational and equipment requirements Incomplete recovery	Reduce recovery costs Lower the requirements for categories Further optimize product performance

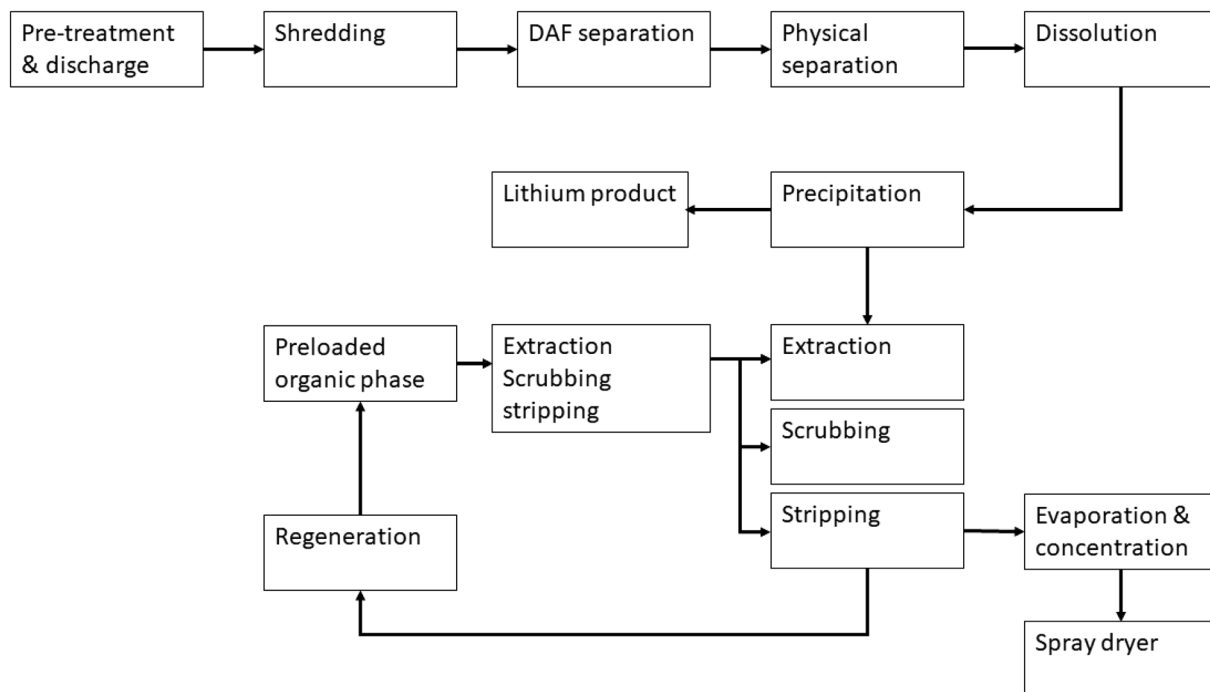


Fig. 10 Proposed hydrometallurgical scheme for LIB recycling process

blowers, the lightweight plastics are segregated from the metals. Further separation processes, including magnetic separation and use of a vibrating screen, are applied to the metal components. Dissolved air flotation is implemented to eliminate low-density shredded elements by diverting them from the tank collector. This precedes a physical separation stage where a cylindrical arrangement of permanent magnets, featuring alternating polarities, is rotated, generating a time-varying magnetic field. Under sufficient magnetic force, nonferrous metals (such as aluminum) and nonmetals (like LFP) detach from the feed belt, subsequently isolated via a mechanical splitter [90]. Next, a dissolution process is initiated to separate the cathode active material (CAM), with any ensuing gas potentially directed to an absorption unit for treatment. The resultant solution is then directed to a mixer settler, comprising three primary stages: extraction, scrubbing, and stripping [74]. The ensuing precipitated lithium product is subsequently transferred to tray dryers for further processing.

Conclusions

LIBs are widely used in electronics, solar panels and most importantly EVs. Due to the anticipation of the increase in the EV demand in the future, it is expected that more waste LIBs will be generated and disposed in landfills, affecting the groundwater and the environment. Thus, recycling LIBs is

needed to lower the environmental impact that can pose imminent risks. Furthermore, a feasibility study was performed and proved that recycling LIBs consumes less material and energy and emits less GHG than producing LIBs from virgin materials. Therefore, this paper extensively reviewed different recycling methods, mainly, hydrometallurgy, pyrometallurgy, direct recycling and precise separation. It also reviewed an additional method of recycling waste LFPs which is the eddy current separation for recovering aluminum and LFP components. Then, a process selection matrix was made and hydrometallurgy was chosen as the most efficient method to recycle waste LIBs on an industrial scale. Although this paper provided good understanding about waste LIBs recycling in general, and hydrometallurgy in particular, it still requires further experimental work to verify the success of hydrometallurgy in recycling waste LIBs.

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Declarations

Conflict of 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.

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