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Recycling lithium-ion batteries: A review of current status and future directions

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ABSTRACT

Lithium-ion batteries (LIBs) have become a widely adopted energy source for various electrical devices, ranging from small devices to large machines, such as cell phones, and electric vehicles (EVs). The increasing number of EVs, and other electrical devices has led to the enormous amount of discarded spent LIBs into the landfill. The amount of LIB waste generated in 2019 alone from EVs was 500,000 tons. This amount is expected to reach 8,000,000 tons by 2040. Globally, only 5 % of discarded spent LIBs is presently being recycled. The need to recycle LIBs stems from the desire to conserve raw materials, and save cost. Also, LIBs comprise heavy metals (Ni, Li, Co, Cu, Mn, Fe, and Al), and hazardous chemicals, which cause serious environmental hazards and threaten human lives; thus, pointing out the need to recycle LIBs. This work reviewed different recycling techniques and, the latest technological advancements in pretreatment methods, hydrometallurgy, pyrometallurgy, and direct recycling methods. Also, future perspectives and prospects are provided herein.

1. Introduction

Batteries store chemical energy, which is converted into electrical energy through electrochemical reactions for use in electronic appliances. Batteries can be grouped into rechargeable and non-rechargeable, depending on the utility [1]. Rechargeable batteries can undergo several cycles of recharge before their end-of-life, and they are listed as follows: Lead-acid batteries, Lithium-ion batteries (LIBs), Nickel-metal hydride (NiMH) batteries, and Nickel-cadmium (Ni-Cd) batteries. Among all, LIBs are leading other rechargeable batteries in terms of easy charging and maintaining, increased energy density, extended life span, higher cell voltage, lower self-discharge, lesser memory effect, and environmentally sound [2]. A battery's core composition (Fig. 1) consists of four primary elements: the positive cathode, the negative anode, the conductive electrolyte, and the separator (i.e., separating membrane) [3]. Other components of LIBs are metal casing (protective), control unit, and covering plastic. The anodes of these batteries comprise carbon graphite. Polyvinylidene fluoride (PVDF) binder acts as a glue holding the carbon graphite active material to the copper plate [4]. The cathode is the crucial component in LIBs and it is typically coated with an aluminum foil, which enhances its performance and durability. Cathodes comprise carbon powder, polyvinylidene binder, and transition metal oxides of lithium like LMO ($LiMn_2O_4$), LCO ($LiCoO_2$), NCA ($LiNi_xCo_yAl_zO_2$), LFP ($LiFePO_4$) and NCM ($LiNi_xCo_yMn_zO_2$). The electrolytes are Lithium salt solutions such as $LiPF_6$, $LiClO_4$, and $LiBF_4$. Certain organic solvents like Dimethyl Carbonate (DMC), or Ethylene Carbonate (EC) are usually used to dissolve the electrolytes [5]. The ions utilize the electrolytes, as a medium, through which they move between electrodes [6,7]. The separator is a non-conductor, made of polymeric materials like polyethylene (PE) or polypropylene (PP) [8]. The essence of having a separator, installed in between the anode and cathode is to act like a barrier, to avoid the direct contact of short-circuiting between the two electrodes.

In small electronic devices, LIBs can last about three years, and about four to ten years in larger devices. The amounts of LIBs utilized in tiny devices are more than 80 %, while less than 20 % are utilized in storage

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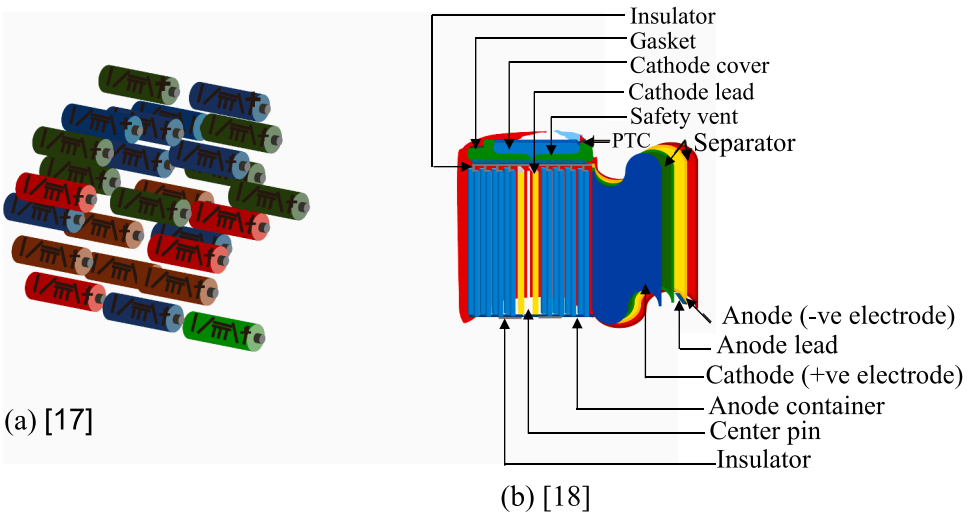


Fig. 1. Lithium-ion battery structure: (a) schematic sketch (b) cell structure.

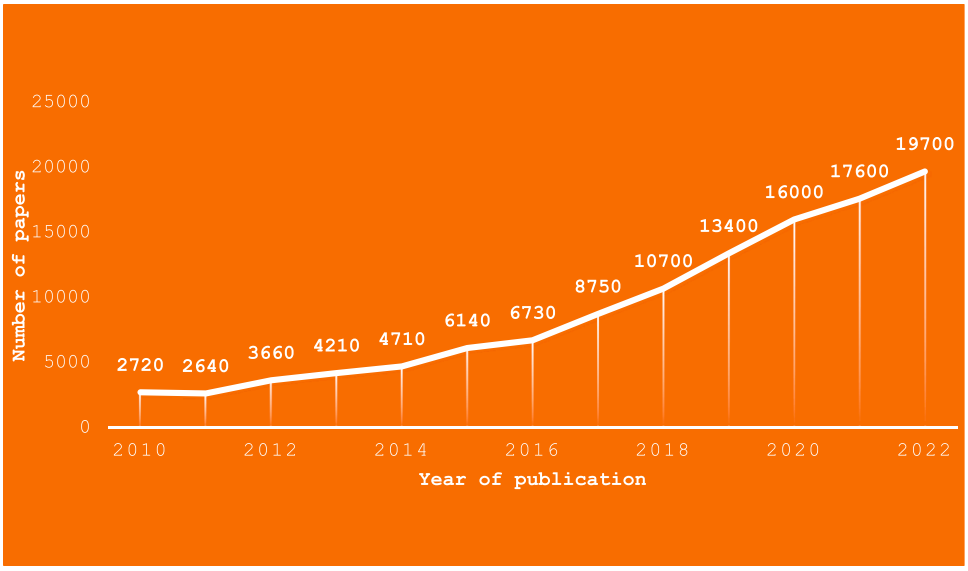


Fig. 2. Volume of published papers about lithium-ion battery recycling [23].

systems and electric vehicles [9]. In 2012, the total estimate of disposed LIBs was about 10,700 tons [10]. The amount has risen annually surpassing an estimated value of 250,000 tons in 2020. Research by Mossali et al. [11] shows that most of the population that use portable electronics devices store the spent LIBs at home, about 59.6 %, or throw them away, about 15.9 %, hence, only a very small fraction of the population properly collects (for recycling) the spent LIBs at the end-of-life span [11]. Recently, an increase in the number of operating EVs has led to an enormous rise in the amount of discarded spent LIBs. The urge to recycle spent LIBs ranges from environmental protection and climate change mitigation to resource conservation and life protection. Spent LIBs contain hazardous chemicals which have the potential to cause severe environmental and atmospheric hazards (such as air pollution from toxic gas emissions, greenhouse gas emissions, particulate matter emissions – Pb, Ni, Cd, Li, Co, Al), and pose a big threat to lives. The spent LIBs are made of substantial amounts of heavy metals (Lithium, Nickel, Cobalt, Copper, Manganese, Iron, and Aluminum) [12]. Another research from Yao *et al.* shows that these metals found in spent LIBs can even be higher in amount than those existing in the natural ores [13]. The percentage by composition of the materials of LIBs differs a little among producers and may include 5 – 7 % of Li, 5 – 20 % of Co, 5 – 7 % of Ni, 7 % of plastics, and 15 % of organics [14]. Therefore, it is important to collect and carefully recycle spent LIBs for resource conservation, environmental and life protection, leading to increase in research in the area of LIB recycling (Fig. 2). Recycling also serves as an essential pathway for the recovery of important strategic elements, especially Li, Co, and rare metals [15]. Tables 1 and 2 present the factors which necessitate the recycling of LIBs. Recycling is the ultimate fate of spent LIB, which is the major thing we are considering in this article. Other management options (which can be categorized as "re-using of spent LIBs") are also available for spent LIBs, which include repurposing and remanufacturing [16]. Repurposing is the reutilization of spent LIBs in other electrical devices that have lower energy demands. The battery modules are reconfigured and damaged cells are also changed to accommodate a second life usage. Remanufacturing involves dismantling of LIB packs, changing of damaged cells and parts of the LIB, and reassembling them into new packs [17,18]. Remanufactured LIB can serve its main purpose or other purposes, such as reutilizing EV batteries in lighting up a bulb. The advantages and disadvantages of repurposing and remanufacturing of spent LIBs are presented in Table 3.

In this review, we comprehensively show the current status of LIBs, factors that necessitate the recycling of batteries, environmental impacts

Table 1
Factors that necessitate the recycling of LIBs [19,20].

Environmental concern of LIB disposal	
Factor	**Material release**
Toxic materials	LIBs contain materials like Co, Li, Pb and Ni which can contaminate soil and water if not recycled.
Water pollution	The release of Co, Li, Pb and Ni into the water bodies causes water pollution, causing harm to aquatic life and human health.
Soil pollution	The hazardous chemicals present in LIBs affect plants growth and food chain.
Greenhouse gas emission	Release of greenhouse gases can vary depending on energy sources and production processes. However, CO_2 and CH_4 are generally released.
Electronic waste accumulation	Discarded LIBs are electronic wastes on their own. They release toxic wastes, cause water and soil pollutions, and release of greenhouse gases.
Loss of valuable materials and resource depletion.	Materials like Co, Li, Pb and Ni are valuable and limited. Not recycling lithium ion can potentially lead to scarcity of resources and environmental degradation.
Economic Benefit of recycling LIBs (Economic Aspects for Recycling of Used Lithium-Ion Batteries from Electric Vehicles).	
Benefits	**Material gain**
Resources conservation	Co, Li, Pb, Ni, and graphite are recovered through recycling, hence reducing reliance on primary extraction
Reduced waste management cost.	When LIBs are recycled, the amount of waste entering the landfill is reduced
Overall cost savings	Recycling LIBs can be cost effective than initial cost of resource mining (raw materials) and extraction

Table 2
Cost-benefit analysis for recycling lithium-ion battery – The economic value of the components of LIB [11].

Components	Battery element	Price (USD per kg)		
		2001	2017	2019
Cathode	Al	1.25	2.00	1.80
	Li	7.50	9.00	10.0
	Co	38.00	55.00	35.50
	Ni	8.60	10.00	13.20
	Mn	1.10	2.00	2.00
Anode	Cu	1.80	5.50	5.80
	Graphite	0.55	1.00	0.80

Table 3
Pros and cons of repurposing and remanufacturing of spent LIBs [21,22].

Repurposing of spent lithium-ion battery	
Pros	**Cons**
Extended life span	Limited compatibility
Lower cost	Reduced performance.
Reduced waste	Potential safety risks when not retested and certified properly
Energy storage for secondary applications	
Remanufacturing of spent lithium-ion battery	
Pros	**Cons**
Reduced material wastes	Energy and resource intensive
Reduced cost than recycling	Higher cost than repurposing
Restored to original performance	Potential for reduced life span
Undergoes thorough testing and certification	Requires advanced technology

of not recycling spent batteries, cost considerations and recycling methods. The recycling methods explained in this article for LIBs are pretreatments, pyrometallurgical, hydrometallurgical, and direct recycling processes. Following on, the comparisons of the recycling methods are evaluated, including their pros and cons to give readers more in-depth research. Finally, future directions are given to illustrate critical perspective and uphold the sustainability of battery industry by defining optimization path for further design and production of batteries.

2. Lithium-ion batteries recycling processes

The three major methods of recycling LIBs are pyrometallurgical, hydrometallurgical, and direct recycling processes. Pyro- and hydrometallurgical processes are chemical processes, while direct recycling is a physical process [24]. Pyrometallurgy is a heat treatment process, under elevated temperature, then followed by separation processes. In Hydrometallurgy, the valuable components of LIBs are dissolved in aqueous solutions and then recovered through the concentration and purification stages. In direct recycling, the LIB's components are recovered directly. Hydrometallurgical and Direct recycling processes may require pretreatment methods. The pretreatment is necessary to access the cathodic black mass and separate Al foils and organic binders from it. The Al foils and organic binders make the black mass hard to leach and also cause complications of the leachate [25]. Hence, pretreatment enhances the subsequent processes of recovering of the cathodic components of the spent LIB, and increases the overall efficiency of the recovering process.

2.1. Pretreatment methods

The pretreatment methods are important in order to separate different elements of the battery and access their active valuable substances. They generally involve three steps (Fig. 3): Discharging, Dismantling, and Separation.

2.1.1. Discharging

Spent LIBs can be discharged by soaking them in a saturated salt solution, like Sodium Chloride, NaCl, or Sodium Sulphate, Na_2SO_4. This minimizes short-circuiting and material deposits in the anode [27]. Another way of discharging Lithium-ion batteries is by connecting them to resistors. By this approach, the residual energy can be extracted and reused, instead of being wasted. A closed circuit is created when a battery is connected to a resistor. This allows electrons to flow out from the negative terminal of the battery, through the resistor, and return to the battery through the positive terminal. The flowing out of the electrons from the battery gradually depletes the chemical energy of the electrolyte thereby draining the battery. This approach is not appropriate for massive battery discharging, as there is a greater recycling cost than gain. Fig. 4 shows the charging and discharging of LIB.

2.1.2. Dismantling or disassembling

Disassembling of batteries can be done automatically or manually [29]. Manually disassembling batteries can be done using hand tools like saws, knives, and pincers. During this operation, the battery casing is removed first before the disassembling of the other parts of the battery. This is done to access the cell core. This technique is not ideal for the mass disassembling of batteries. The mechanical separation process, which has economic and industrial advantages, is the optimum operation for the mass disassembling of scrap batteries [30].

2.1.3. Separation

The separation is necessary before embarking on the Hydrometallurgy process to eliminate impurities from the aluminum and copper foils, as these obviate the leaching process. The techniques commonly used in separating components of the scrap batteries are physical and mechanical, thermal, chemical, and mechano-chemical separation processes.

2.1.3.1. Physical and mechanical separation. This treatment process is highly dependent on the physical properties of the scrap batteries, such as size, shape, and material type. The separation includes crushing,

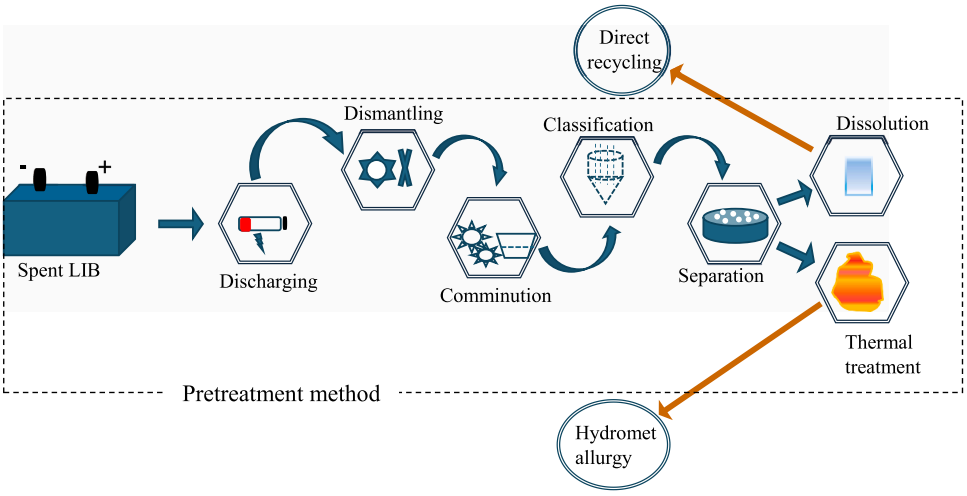
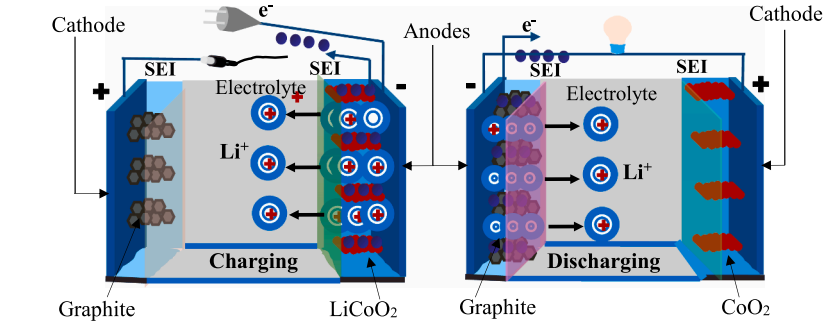


Fig. 3. Overview of pretreatment recycling method [26].



Reduction happens at the Cathode:
$C_6 + e^- + Li^+ \rightarrow LiC_6$
Oxidation happens at the Anode:
$LiCoO_2 \rightarrow CoO_2 + e^- + Li^+$

Oxidation happens at the Anode:
$LiC_6 \rightarrow C_6 + e^- + Li^+$
Reduction happens at the Cathode:
$CoO_2 + e^- + Li^+ \rightarrow LiCoO_2$

Fig. 4. Charging and discharging of Lithium-ion cells [28].

sieving, magnetic separation method, eddy current separation, and flotation. This method follows after the dismantling of the battery. The sieving separates the current collector and organic materials from the active leachable powder [31].

Crushing: The two techniques used during crushing are wet and dry processes. In wet crushing, the spent LIB is ground in the salt solution, after self-discharging in the same solution. Salt solutions such as NaCl, $MgSO_4$, and Na_2S can be utilized in wet-crushing [32]. This process prevents the emission of toxic gases and the reactiveness of lithium [33], and may lead to loss of fine particles due to the flowing action of water [34,35]. Dry crushing of spent LIBs is carried out in an inert atmosphere, where there are no risks of contamination by reactive gas. This is fit from practical and safety viewpoints. Dry crushing can optimize the productivity and effectiveness of the downstream processes [36]. It also prevents short-circuiting and reduces the risks associated with impact crusher. Shredders, cutting mills, granulators, and impact crushers are the most inspected crushers. Impact crushers and shredders also have been used in several investigations [37], because fine particles may jump out in granulators due to their high speed, as the centripetal force which keeps the fine particles together tends to reduce as the particle size reduction process continues. Crushing of a battery can be accomplished in one step or more. An example of a two-step process is the 'Recupyl process', here the high-speed impact crusher, which is way above the first crushing, is used to accomplish the second crushing [38]. Crushing twice remarkably enhances the black mass yield to about 15 % increase, and reduces the loss of materials to about 4 % in the subsequent separation processes [39,40]. One drawback in the second crushing is that this may increase the impurities in the black mass, like Aluminum and Copper foils. Hence, proper control needs to be given to the crushing time. Granata examined the recovery efficiency of a two-step crushing performance and the recovery efficiency of the combination of a two-blade rotator crusher which is then followed by a hammer crushing. Comparing the two methods, it was discovered that more electrode powders were recovered in the latter than in the former. Increasing the surface area of the crushed products allows for greater exposure to acid, leading to improved metal solubility and more effective extraction during the acid leaching of the hydrometallurgical processes [41].The particles of the plastics, aluminum, and copper foils appear in coarse form after crushing [42], while those of the graphite and electrode materials like $LiCoO_2$ appear in fine forms.

Sieving: Sieving as a separation technique depends highly on the particles' size differences. Sieving is utilized to remove the electrode materials (known as the fine particles, called the black mass) from the other components (known as the coarse particles) of the LIB [43]. Sieving can also be done using an automatic screening system [44]. The sizes of both the fine and coarse classifications differ slightly according to the LIB type. This can range from 0.5 mm to 63 mm [45]. An excellent comminution circuit (efficient crushing, then screening) can be used to remove the Al-Cu foils from the electrode powder. This has been seen to reduce the required processes of hydrometallurgy in the extraction of

Table 4
Conditions for thermal treatment in recycling spent LIBs (All experiments were conducted on the laboratory scale).

Operating conditions of lab scale thermal treatment		Particle size (mm)	Reference
Conditioning Temperature (℃)	Conditioning Time (hour)		
Anode 550	0.25	<0.2	[46]
500	0.5	0.075	[59]
500	0.16	<0.2	[60]
1000	0.5	<0.12	[61]
450	0.25	<0.25	[62]
500	2	0.25	[63]
400	1	<0.1	[64]
800	0.33–0.5	<0.12	[65]
800	0.75	<0.12	[66]
750	4	<0.074	[67]
700	5	<0.106	[68]
700	0.5 – 2	<0.063	[69]
700	5	<0.1	[70]
700	2	<0.075	[71]
400 – 700	0.5 – 2	<0.1	[72]
650	5	<0.05	[73]
550 – 650	0.25	<0.1	[74]
600	3	0.01	[75]
600	1	<0.045	[76]
450	2	<0.2	[77]
150 – 600	2 – 3	1 – 5	[78]
550	1	<0.075	[79]
500	1	10	[80]
500	2	<3	[81]
500	3	<8	[82]
500	0.5	0.002 – 0.01	[83]
150 – 500	1	0.00014	[84]
400	2	<0.2	[85]

valuable metals.

Magnetic Separation: The components of the crushed batteries containing iron (like the Al-Cu electrodes and steel casing) are isolated from other components by Magnetic separation [46,47]. Li and his research team employed wet magnetic separation to process a blend of virgin cathode materials, separating and purifying the components [48]. Another research from Sommerville and his research team revealed that more than 90 % of recoveries of each fraction could be received [49]. Qiu and his colleagues proposed an environmentally-friendly separation method for fluid-magnetic separation. This is used for recovering cobalt and nickel ferromagnetic metals from the LIBs vacuum reduction products. The rates of recovery were twice more than the normal magnetic separation [50]. 98.26 % recovery rate for the corresponding magnetic metal particles was achieved, with 99.21 % for cobalt and 97.91 % for nickel.

Eddy Current Separation: Al and Cu foils are isolated from non-metals (>1 mm) and nonferrous metals by the use of eddy current separation method [51]. The intensity energy generated by the Al and Cu foils in an alternating magnetic field is very high due to their proper electroconductivity [52]. Yamaji and his research team explored the application of eddy current to recover Al and Cu foils (-4.8+1.0 mm), occurring at a speed belt of 0.78 m/s. Eddy current/Electrostatic separation was also utilized for fractions of finer sizes, at lower speed belt to increase efficiency of separation [53].

2.1.3.2. Thermal separation. Heat treatment is used to remove bonds of organic binders like Polyssvinylidene Fluoride (PVDF) from cathodes, and separate the active materials [54,55]. The separation efficiency of the spent LIB would be low if thermal treatment is neglected. Thermal treatment of various size fractions will be wisely considered since it could be done for varied purposes [56]. The Table 4 presents the consideration of particle size fractions in thermal treatments. It is quite inappropriate that a lot of investigations didn't report this. Lee and Rhee originally treated LIB samples for 30 mins, at a temperature ranging from 100 to 150℃. The treated samples were disassembled, and crushed, and the crushed samples were again subjected to thermal treatment for 1 hour at a temperature ranging from 300 to 500℃. The current collectors liberated were separated from the electrode materials, and the resulting $LiCoO_2$. Then the organic binders and their bonds burned at a temperature ranging from 500–900℃ for about 0.5–2 h [57]. This treatment referred to as pyrolysis, releases some dangerous gases, known as pyrolytic gases into the atmosphere. Pyrolytic gases usually contain light alkenes, methane, carbon monoxide, hydrogen, and other hydrocarbons. Pyrolytic tar is another product of pyrolysis. Tar usually has light alcohols and long-chain aromatic alkenes. It can be reutilized as a fuel in pyrolysis operation, thereby minimizing consumption of energy and preventing pollution of the environment. After completion of the pyrolysis, the organic materials were evaporated or reduced to products of lower molecular weight, while the inorganic components, at the same time, remained as solid residues. The organic materials are recovered during pyrolysis by making the pyrolytic gases pass through lines of condensers and scrubbers. All the different constituents of the organic material available in the pyrolytic gases will be captured and separated using this format [58].

2.1.3.3. Chemical separation. Alkaline solutions and organic solvents are used in chemical preliminary separation treatment to free the electrode substance from aluminum foils. An organic solvent like N-methylpyrrolidone (NMP) is used usually because the solvent is polar, just like the polyvinylidene fluoride (PVDF) binder, thereby allowing them to go into a solution with each other. According to research, additional organic solvents that can also be used to separate the cathodic active substances are ionic liquid [86], N, N-dimethylacetamide (DMAC) [87], dimethylsulfoxide (DMSO) [88], and N, N-dimethylformamide (DMF) [89]. According to Li and his colleagues, the active substances of the cathode can be detached from the PVDF binder by immersing it in NMP at a temperature of 100℃ for 1 hour [90]. Yao and his colleagues, in their study, first immersed the positive electrodes in NMP at a temperature of 80℃ for 1 hour, and then treated using ultrasonic, which was found to increase the effectiveness of the process and also the separation of the aluminum sheet from the positive electrode substances [91]. Aluminum current collectors are recovered in metallic form during the dissolution process. The choice of organic solvent is strictly binder-specific, as different binders require distinct solvents, and there is no universal solvent that can effectively dissolve all types of binders. N-methylpyrrolidone is not effective for binders that are made of polytetrafluoroethylene (PTFE) due to their non-polar nature, which makes them resistant to dissolution by this solvent. Instead, they work well on PVDFs. Also, calcination processes may be employed to burn off all the residuals of PVDF or Carbon (C), because the organic solvents could not eliminate the impurities. The dissolution reagent is expensive, poisonous, and not suitable to apply in large-scale processes. A green citrus fruit juice was utilized as a dissolution solvent reagent. This is to prevent the environmental downside associated with harmful reagents [92]. But using citrus fruit in the dissolution process has to be done at a high-temperature of over 90℃. Alkaline solutions (NaOH) are efficiently employed to separate active materials because they don't react with the cathodic oxide materials. Alkaline solutions are used in dissolving Aluminum layers, then their binders are removed through processes of calcining. This is done to have a pure electrode active substance [93]. Though this process has a high efficiency in extracting aluminum and other metals preservation, losses are unavoidable. Also, Alkaline solution dissolution presents impurities that hinder subsequent processes of materials recovery [94].

2.1.3.4. Mechano-chemical separation. A pretreatment method that combines both the mechanical and chemical methods in separating the active substances of the LIBs is known as the Mechano-chemical technology. The benefit of mechanical shredding as well as chelate reagents

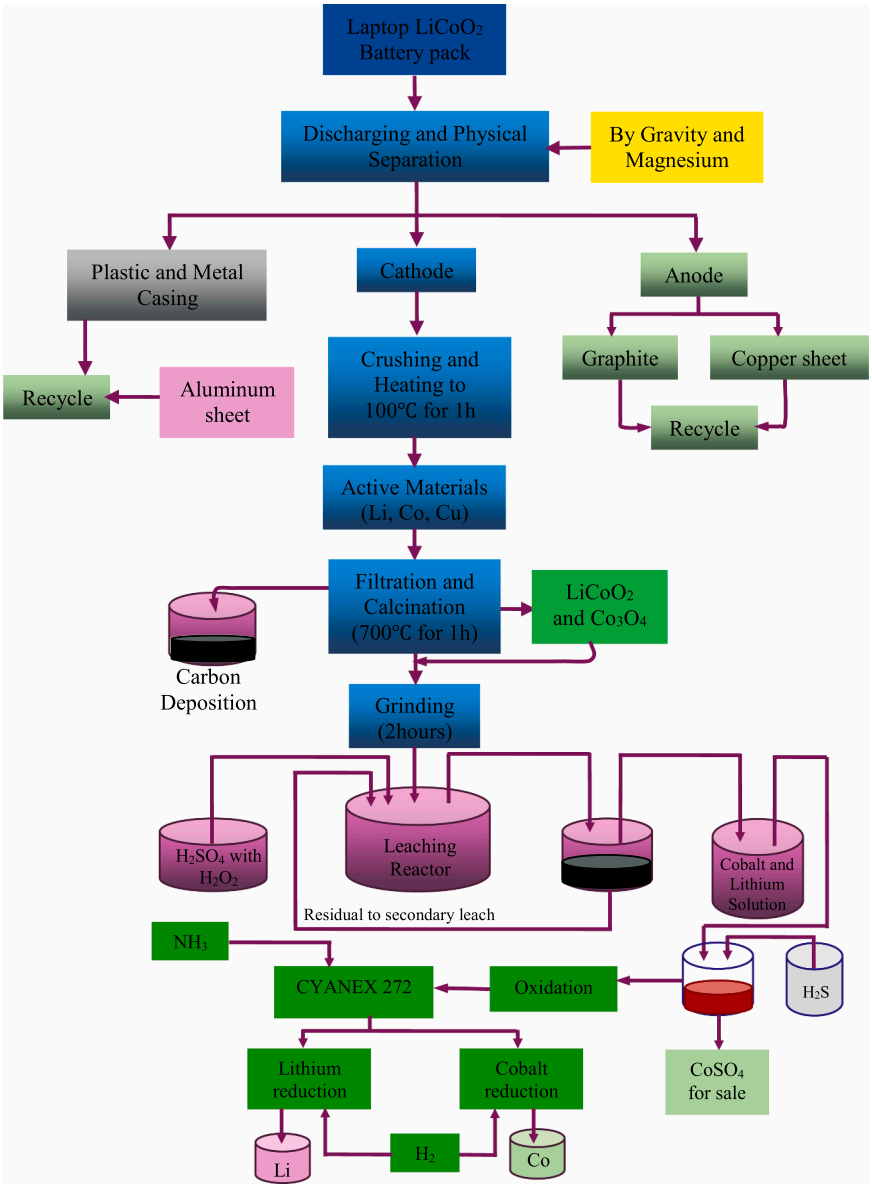


Fig. 5. Hydrometallurgy flow diagram [108].

is utilized in this method. Chelate reagent like Ethylenediaminetetraacetic (EDTA) is used. The bond breakage and the polymorphic transformations of cathode substances, which lead to particle size lessening and surface area enhancement, make it possible for the process to pave the way for subsequent acid-leaching processes [95]. The major drawback of this method is its long reaction time, which makes it hard to use in industries [96].

2.2. Hydrometallurgy

After pretreatment methods, hydrometallurgy is used to recover valuable components of the Cathode materials. Hydrometallurgy involves leaching and reduction processes. The leaching methods used are acid leaching and biological leaching methods. A report from Zhang and his colleagues shows more than 99 % recovery rate for Co and Li, in the acid-leaching process [97], while another report from Nan and his colleagues shows a more than 98 % recovery rate for Cu [98]. Inorganic acids that are typically employed in the acid leaching of electrode materials include Sulfuric acid (H_2SO_4) [99], Hydrochloric acid (HCl) [100], Phosphoric acid (H_3PO_4) [101], and Nitric acid (HNO_3) [102, 103], Some of the organic acids used include Oxalic acid [104], Tartaric acid, and Citric acid [105,106], The principle of acid leaching using HCl to recycle $LiCoO_2$ was shown by Lee and his research team [107]as follows:

$$2LiCoO2 + 8HCl \rightarrow 2CoCl2 + Cl2 + 2LiCl + 4H2O.$$

During the process, the acid leaches Co from the spent LIBs and reduces the Co_3^+ to Co_2^+. A Cl_2 is also produced during the process. The presence of this gas is a big drawback because it's very corrosive and toxic. One of the optimum ways of solving this problem is by using sulfuric acid (H_2SO_4) for the hydrolysis (Fig. 5). Then, the Co_3^+ formed is reduced to Co_2^+ using hydrogen peroxide (H_2O_2). The reaction follows thus:

$$2LiCoO_2 + 3H_2SO_4 + H_2O_2 \rightarrow 2CoSO_4 + O_2 + Li_2SO_4 + 4H_2O.$$

In acid leaching of NCM ($LiNi_xCo_yMn_zO_2$) electrode materials, strong acidity HCl should not be used. This is to lessen Mn dissolution. A report from Castillo and his research team on acid hydrolysis of NCM cathode materials using nitric acid showed recovery rates of 100 % Li and 95 % Mn [109]. Physical quantities like pH value, reaction time, temperature,

Table 5
A comprehensive summary of previous studies on hydrometallurgical process.

Spent cathode	Recovery efficiency	Leaching agent	Reducing agent	Critical operating factors		References
				Temp, ℃	Residence time (h)	
LCO	Co (>99 %), Li (>99 %)	2 M H_2SO_4	5.0 vol% H_2O_2	80	1	[58]
LCO	Co (98.5 %), Li (99.8 %)	2 M H_2SO_4	10 vol% H_2O_2	70	1	[121]
LCO	Co (98.3 %)	2 M H_2SO_4	H_2O_2	25	4	[122]
	Co (98.4 %)			50		
LCO	Co (99 %), Li (98 %)	5 M HCl	-	95	$1\frac{1}{6}$	[123]
LCO	Co (>89 %), Li (>89 %)	3 M HCl	3.5 vol% H_2O_2	80	1	[124]
LCO	Co (99 %), Li (99 %)	1 M HNO_3	1.7 vol% H_2O_2	75	0.5	[107]
LCO	Co (99 %), Li (88 %)	H_3PO_4, 2 % v/v	2.0 vol% H_2O_2	60	1.5	[101]
LCO	Co (96.3 %), Li (87.5 %)	2 M H_2SO_4	2.0 vol% H_2O_2	60	2	[15]
LCO	Co (70.0 %), Li (99.1 %)	2 M H_2SO_4	5.0 vol% H_2O_2	75	1	[111]
LCO	Co (98 %), Li (97 %)	2 M H_2SO_4	6.0 vol% H_2O_2	60	1	[125]
LCO	Co (99.7 %), Li (99.9 %)	0.7 M H_3PO_4	4.0 vol% H_2O_2	40	1	[126]
LCO	Co (80 %), Li (95 %)	6 vol% H_2SO_4	5.0 vol% H_2O_2	65	1	[127]
LCO	Co (93 %), Li (94 %)	2 M H_2SO_4	5.0 vol% H_2O_2	75	0.5	[128]
NCM	Co (90.9 %), Li (91.0 %)	0.5 M citric acid	-	90	$1\frac{1}{3}$	[129]
LCO	Co (99 %), Li (99 %)	2 M citric acid	0.25 M H_2O_2	80	2	[130]
LCO	Co (97 %), Li (89 %)	1 M citric acid	1.0 vol% H_2O_2	RT	24	[131]
LCO	Co (98 %), Li (96 %)	1.5 M citric acid	2.0 vol% H_2O_2	95	0.5	[132]
LCO	Co (90.2 %), Li (98 %)	1.25 M citric acid	0.9 vol% H_2O_2	90	$\frac{35}{60}$	[133]
LCO	Co (90.2 %), Li (98 %)	2 M D,L-malic acid ($C_4H_5O_6$)	6 vol% H_2O_2	95	1	[134]
LCO	Co (47.6 %), Li (97.2 %)	0.5 M H_2SO_4	0.55 M H_2O_2	60	5	[90]
LCO	Co (76.4 %), Li (95 %)	0.5 M HCl				
NCM	Co (96.1 %), Li (98.4 %)	0.5 M citric acid	0.55 M H_2O_2			
LCO	Co (90.2 %), Li (98 %)	0.1 M citric acid	0.02 M ascorbic acid	80	6	[135]
LCO	Co (90.2 %), Li (98 %)	0.4 M tartaric acid	0.02 M ascorbic acid		5	[136]
LCO	Co (90.2 %), Li (98 %)	0.1 M iminodiacetic acid	0.02 M maleic acid + ascorbic acid		6	[126]
LCO	Co (90.2 %), Li (98 %)	0.5 M glycine	0.02 M ascorbic acid			[137]
LCO	Co (87.85 %), Li (99.91 %), Ni (1.46 %)	1.5 M H_2SO_4	30 % H_2O_2/glutaric acid	90	2	[138]

and additives, all have effects on the performance of inorganic acids during the leaching process. Li and his colleagues (2009) leached a 4 M HCl solution for 2 hours at 80℃, 97 % Li and 99 % Co were dissolved [110]. Jha and his colleagues (2013) reported the leaching performance of 2 M H_2SO_4 and H_2O_2 5 % V/V, on $LiCoO_2$ for 1 hour at a temperature of 75℃, and achieved 99.1 % Li and 70 % Co dissolutions [111]. A report by Chen and his colleagues (2017) showed the separation of precipitate from a lithium-ion containing solution [112], while Shih and his colleagues (2019) studied the leaching performance of H_2SO_4 on $Co_3(PO_4)_2$ in the presence of microwave and ultrasonic at 90℃, and reported that Co was completely dissolved and average of 90 % of the total metals present were leached [113]. Lee and his 2003 research team revealed that treating $LiCoO_2$ with a solution of 1 M HNO_3 and 1.7 % V/V H_2O_2 for 30 minutes achieved an impressive 95 % dissolution rate for both lithium and cobalt at a temperature of 75℃. Phosphoric acid is a viable alternative to strong inorganic acids for acidolysis, providing a viable solution for this chemical process. According to 2017 research by Pinna and his research team, treating $LiCoO_2$ with a solution of 0.7 M H_3PO_4 and 4 % V/V H_2O_2 at 40℃ for 1 hour achieved a recovery rate of over 99 % for both Li and Co.

During oxalic acid leaching, the processes of leaching and precipitation occur simultaneously, resulting in the formation of CoC_2O_4 precipitate, which can be directly separated from the lithium-ion solution without additional treatment. The reductive nature of the oxalic acid renders additional reductants unnecessary. According to Sun and Qiu's 2012 study, a recovery efficiency of more than 98 % for Li^+ and Co_2^+ ions, achieved through a 2-hour treatment with 1 M acetic acid was reported, demonstrating the effectiveness of this leaching process [114]. A research paper by Chen et al. in 2016 investigated the leaching capabilities of citric acid, shedding light on its potential as a leaching agent for various applications. A leaching process using citric acid was applied to spent $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ electrode material at 80℃ for 2 hours, yielding a high recovery rate of lithium (99 %), nickel (91 %), cobalt (93 %), and magnesium (94 %) [115]. In a separate study, Chen and his research team, in 2019, utilized tartaric acid to achieve an excellent recovery performance of 97 % of lithium and 98 % of Cobalt from $LiCoO_2$, resulting in the formation of a precipitate and Lithium enriched solution[116].

In the biological leaching method of the hydrometallurgical process, microorganisms like fungi and bacteria are used in the extraction of valuable metals from spent electrode materials. By secreting metabolites, these organisms facilitate the dissolution and release of the metals, making them accessible for recovery and reuse. The availability of slurry concentration affects the bioleaching; thus, the survival of the microorganisms is considered. A research paper by Mishra et al. in 2008 investigated the treatment of $LiCoO_2$ by acidophilic and chemotrophic bacteria [117]. According to Niu et al., 2014, the leaching efficiency of Alicyclobacillus SP for lithium and cobalt decreased significantly as the ore concentration increased from 1 % to 4 %. Specifically, the leaching rate for lithium dropped from 52 % to 10 %, while that of cobalt decreased from 80 % to 37 % [118]. The performance of Aspergillus Niger was reported by Mohagheghi and his 2017 research team [119]. Research by Bahaloo-Horeh and Mousavi in 2017 also revealed similar findings from experiments on Aspergillus Niger as well as the impact of electrolytes in the slurry [120]. A systematic review of previous studies on hydrometallurgy was conducted (Table 5**)** to fully understand the critical operating factors of the process.

2.3. Pyrometallurgy

Pyrometallurgy is used commercially to recover valuable components of the LIBs, using a treatment method that functions closely to the smelting of ore [139]. The process begins with the disassembly of spent LIBs into their constituent components which are then fed into the heating furnace for smelting, marking the first stage of the recycling process. The battery undergoes a multi-stage thermal treatment process involving preheating, pyrolysis, and smelting, which successively breaks down the battery's components, resulting in a complete reduction [140].

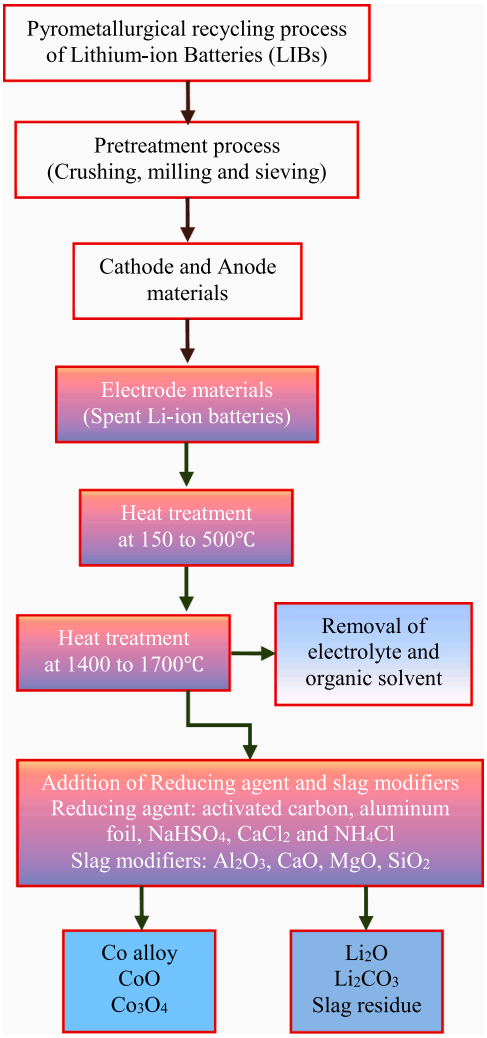


Fig. 6. Pyrometallurgy flow diagram [143].

The preheating is done at a temperature less than 300°C, so as to avoid explosion while ensuring the electrolyte is completely evaporated. The temperature of the furnace is kept above 700°C. This removes the plastic attached to the battery. In the smelting reduction zone, the material is melted to produce alloys of Co, Ni, Cu, and Fe, along with a slag containing Li, Al, Si, Ca, and some Fe. This process usually recovers significant amounts of Cu, Co, Ni, and small quantities of Fe, while the remaining elements are not typically recovered. Because Co is an essential component of LIBs, the profitability of recovering cobalt depends on the quantity of Co in the used LIBs and its market value. The environmental concerns associated with cobalt have led battery manufacturers to consistently develop electrode materials that don't contain cobalt, such as $LiFePO_4$ and $LiMn_2O_4$, with some already being used in commercial applications [141,142]. Taking into account the limited availability of lithium resources, there is no potential for further advancement of cobalt-free electrodes.

The arc furnace pyrolysis process converts spent LIB electrodes into a Co-Li alloy, which is then treated with hydrometallurgy to extract the lithium. The resulting lithium is stored in the form of Li_2CO_3, a valuable resource for reuse. Other components extraction is also carried out further. Fig. 6 shows the steps. The process increases recovery efficiency by not only recycling the electrode components but also recovering lithium and iron from the electrolyte, thereby maximizing the extraction of valuable materials.

Asadi and his colleagues carried out a laboratory-scale investigation in 2020 to evaluate the environmental impacts and economic feasibility of cobalt oxalate produced via acid leaching and roasting techniques and presented their results in a research article [144]. The processing capacity of hydrometallurgy is about 10,000 tons, and it is mainly used in recovering of Li_2CO_3 and Co_3O_4. Anwani and his associates (2020a) produced a study for the evaluation of the life cycle and economic analysis for cobalt oxalate at the laboratory level using acid leaching and roasting techniques [79]. The recovery of cobalt oxalate from 3 g of untreated positive electrode materials was found to incur costs of $0.59 and $0.67 for the acid and baking processes, respectively. Another cost calculation for treating LIBs using acid leaching was given by Anwani and his team in 2020 (i.e., 2020b). This revealed that processing 10 g of material with 250 milliliters of oxalic acid and acetic acid would result in costs of $1.06 and $2.07, respectively [145]. The analysis revealed that oxalic acid was the most cost-effective option, while the most expensive was acetic acid. Pyrometallurgy can cause high consumption of energy, release of toxic gases, such as furans, dioxins, etc, and low recovery efficiency. A process development with low consumption of energy, environmentally friendly, and a high rate of recovery has a better commercial view and should be implemented. We systematically reviewed previous pyrometallurgy researches (Table 6**)** to determine critical process parameters.

2.4. *Direct recycling*

In direct recycling, useful components of the LIBs are recovered without the use of chemicals [162]. After discharging and disassembling (into thousands of cells) pretreatment methods, supercritical CO_2 was used to treat the small cells, and the electrolytes extraction and treatment were also carried out in this process. Lowering the pressure and temperature allows CO_2 to be extracted from the electrolytes. Fig. 7 shows the steps. Chen and colleagues (2016) reported on a technique for recycling $LiFePO_4$ straight from soft-pack batteries [163]. If no electrolytes could be recovered, the LIBs were dismantled, crushed, and cleaned in a sealed environment. The resulting $LiFePO_4$ exhibited suboptimal electrochemical properties due to degradation from repeated cycling and residual polyvinylidene fluoride (PVDF) binder. However, heat treatment at 650°C successfully revitalized its performance, matching the energy density and discharge capacity of fresh $LiFePO_4$ material.

Song and his research team in 2017 developed a novel method to recover $LiFePO_4$ from spent LIBs, yielding a high-quality material through physical direct recovery and sintering [165]. The resulting $LiFePO_4$ battery has electrochemical performance, meeting the basic requirements for reusability. A 2018 research study by Huang and his team investigated three direct physical approaches for recovering graphite anode materials from used LIBs [166]. The first process was heat treatment of graphite without recovering the electrolytes. The second method was to extract the electrolyte and heat it using subcritical CO_2. The third method involved using supercritical CO_2 as an extraction agent along with heat treatment and electrolyte extraction.

The third process is the most effective recuperation method, according to experimental results. Zhang and his research team demonstrated that the cathode materials' liberation efficiency through physical recovery and direct pyrolysis was 98.23 % [167].There is no guarantee that the material recycled by direct method will possess the durable characteristics of the original material. A systematic review of previous studies on direct recycling was conducted (Table 7**)** to fully understand the critical operating factors of the process. The benefits of this recycling system include low energy usage, high recovery rates, environmental friendliness, and a short recovery time.

Table 6
A comprehensive summary of previous studies on pyrometallurgical process.

Pyrometallurgy technique	Cathode material	Additive	Pretreatment	Thermal treatment	Separated material	Secondary process	Recovery rate	Reference
Smelting	Mixed spent LIBs	SiO_2	Roasting at 800℃ for 2 h	1550 ℃ for 0.25 h	Co-Ni-Cu-Fe-Mn and manganese rich slag containing lithium	Acid leaching (H_2SO_4)	Cu (98.67 %), Co (99.84 %), Ni (99.77 %)	[146]
Smelting	LCO ($LiCoO_2$)	Cu slag	-	1450 ℃ for 0.5 h	Co, Ni, Cu, and Fe alloy and slag FeO, SiO_2, Al_2O_3, CaO, MgO.	Manual separation of slag and alloy communtion	Cu (93.57 %), Co (98.83 %), Ni (98.39 %)	[147]
Smelting	NCM ($LiNiMnCoO_2$)	Pyrolusite slag (SiO_2, CaO)	Roasting at 800℃ for 2 h to remove carbon	1475 ℃ for 0.5 h	Co-Ni-Cu-Fe alloy and lithium-containing manganese rich slag	Manual separation, communtion and acid leaching	Li (79.86 %), Mn (94.85 %)	[148]
Reduction Roasting – Thermal reduction	NCM ($LiNiMnCoO_2$)	Carbon	-	650 ℃ for 0.5 h	Li_2CO_3, Co, Ni, NiO, MnO, CO_2(g)	Water and acid leaching (H_2SO_4)	Li (93.67 %), Co (98.08 %), Ni (93.33 %), Mn (98.68 %)	[149]
Reduction Roasting – Thermal reduction	NCM ($LiNiMnCoO_2$)	Carbon	Alkali leaching, 1.5 mol/L NaOH	650 ℃ for 3 h	Li_2CO_3, $NiSO_4$, $CoSO_4$, and $MnSO_4$	Water and acid leaching (H_2SO_4)	Li (84.7 %), Co (>99 %), Ni (>99 %), Mn (>99 %)	[150]
Vacuum pyrolysis –Thermal reduction	LCO ($LiCoO_2$)	Carbon	NaCl discharging, Manual dismantling, Vacuum pyrolysis	600 ℃	CoO, Co, Li_2CO_3, CO_2(g)	Water leaching	Li (93 %), Co (99 %)	[151]
Plasma Spray Pyrolysis – Thermal reduction	NCM ($LiNiMnCoO_2$)	-	NaoH dissolution	600 ℃	Regenerated $LiNiMnCoO_2$	-	-	[152]
Microwave Carbothermic Reduction – Thermal reduction	NCM ($LiNiMnCoO_2$)	Carbon	NaCl discharge, Manual dismantling, Comminution	900 ℃, 500 W, 0.5 h	-	Acid leaching (HCl (aq))	Li (99.68 %), Co (97.85 %), Ni (97.65 %), Mn (96.73 %)	[153]
Oxygen-free roasting – Thermal reduction	LCO ($LiCoO_2$)	Carbon	-	100 ℃, 0.5 h	Li_2CO_3, CO	Wet magnetic separation	Li (98.93 %), Co (95.72 %)	[61]
Thermal reduction	NCM ($LiNiMnCoO_2$)	-	-	Stage 1: 691℃. Stage 2: 873℃.	Ni metal Co metal	Magnetic separation	-	[154]
Thermal reduction	NCM ($LiNiMnCoO_2$)	-	Dismantling and crushing	520℃, 1 h	$LiAlO_2$, NiO, CoO, and MnO	Alkaline leaching (NaOH) and Acid leaching (H_2SO_4)	Li (99.78 %), Co (99.29 %), Ni (98.62 %), Mn (99.91 %)	[155]
Thermal reduction	LCO ($LiCoO_2$)	-	Crushing	600℃, 1 h	Li_2O, $LiAlO_2$, and CoO	Alkaline leaching (NaOH)	Li (93.67 %), Al (95.59 %)	[156]
Chlorination calcination – Salt roasting	LCO ($LiCoO_2$)	NH_4Cl	Discharging, Manual dismantling, NaOH dissolution	350℃, $\frac{1}{3}$h	LiCl, $CoCl_2$, H_2O(g), Cl(g), N_2(g), NH_3(g)	Water leaching	Li (99.18 %), Co (99.3 %)	[157]
Chlorination calcination – Salt roasting	NMC ($LiNiMnCoO_2$)	NH_4Cl	Dismantling and crushing	350℃, 0.5 h	NH_4MCl_3	Water leaching	Li (>98 %), Co (>98 %), Ni (>98 %), Mn (97 %)	[158]
Sulfation Roasting – Salt roasting	LCO ($LiNiMnCoO_2$)	SO_2 (g)	-	700℃, 2 h	Li_2SO_4, $Li_2Co(SO_4)_2$, CoO, O_2(g)	Water leaching	Li (99.5 %), Co (17.4 %)	[159]
Sulfation Roasting – Salt roasting	NMC ($LiNiMnCoO_2$)	Na_2SO_4	NaCl submersion, Manual dismantling, Calcination	750℃, 1.5 h	Li_2SO_4, MnO, NiO, CoO, CuO_2	Water leaching	Li (85.43 %), Co (84.93 %), Ni (84.93 %), Mn (84.93 %)	[160]
Nitration Roasting – Salt roasting	LCO ($LiCoO_2$)	HNO_3	Mechanical pretreatment	250℃, 1 h	$LiNO_3$, $Co(NO_3)_2$, NO(g), O_2(g), H_2O (g)	Water leaching	Li (93 %), Co (92.9 %), Ni (92.9 %), Mn (92.9 %)	[161]

3. Comparison between hydrometallurgy, pyrometallurgy, and direct recycling

Hydrometallurgy is based upon leaching and reduction processes to recover components of the spent LIBs [172]. It's known for its high recovery rate (over 99 % for Co and Li), high purity of product, low consumption of energy, and is typically used for any type of LIBs (Fig. 8). However, it requires strong inorganic acids which can be corrosive and toxic. The major disadvantages include a long recovery route and the generation of a significant quantity of wastewater (acidic waters, effluents, heavy metal-containing wastes), which brings in water treatment challenges. Other wastes associated with hydrometallurgy are residues, sludges, and gases. Pyrometallurgy on the other hand is a smelting and refining processes of recovering components of the spent LIBs [173]. It is a simple operation and short-flow process which has been utilized commercially. Its limitations include the release of toxic gas, high consumption of energy, and low recovering efficiency (Fig. 8 also). Other wastes associated with pyrometallurgy are: slag, particulate matter flue dust and wastewater. The major challenges are: minimization of energy consumption, environmental hazards and pollution

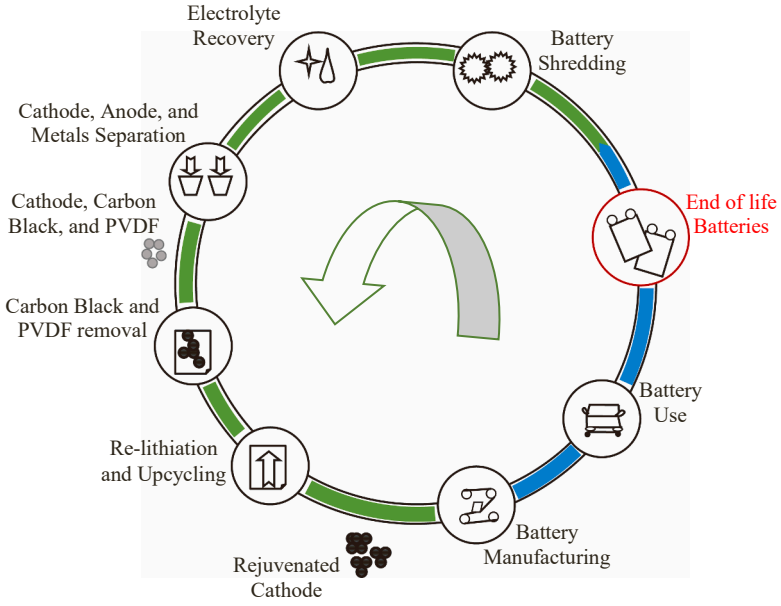


Fig. 7. Direct Recycling flow diagram [164].

emissions [174]. Direct Recycling recovers components of the spent LIBs without chemicals [43]. It has a low consumption of energy, a high recovery rate, short recovery route and is environmentally friendly (Fig. 8). The disadvantages include high cost of operation and equipment requirements, and incomplete recovery of valuable metals. Challenges here are: reduction of recovery costs and optimization of product performance [175]. Direct recycling may release some waste products such as residual metals, contaminated materials and packaging wastes.

The cost comparison analysis of the three recycling methods (Figs. 9**a to** 9**c**) shows the cost variation. Pyrometallurgy has been utilized industrially, and thus requires more capital investment **(**Figs. 9**a to** 9**c).** Another research by [177] shows that direct recycling has more prospect for profit margin (Table 8).

4. Future directions

- The chemistry of battery design and constituents is a complex analysis. Spent batteries are often mixed together, with each varied slightly in design and percentage chemical compositions with one another. The battery sorting, dismantling and separation depend on the material design, internal battery structure and constituent composition. The overall process is tedious. In that note, to make recycling easier, batteries should be designed with recyclability in mind [178]. Intelligence assisted system should be embedded into modern electric vehicle batteries to enhance tracing of battery components and sorting. The pretreatment methods will undoubtedly benefit from this approach. The intelligence system would have the potential to identify optimal disassembling and sorting techniques. Also, classify battery chemistries and materials of the battery. The battery state of health and the remaining capacity can also be determined prior to disassembling. By employing this technique, recycling can be optimized, and the overall efficiency improved.
- Pyrometallurgy is a great industrial technique of recycling lithium-ion battery. However, the quality of the recovered products is poor compare to those from hydrometallurgy and direct recycling [176]. The development of a more efficient pyrometallurgical method will also have a greater advantage from the economic point of view. There are possibilities of plastics contaminating metal alloy or slag if they are not removed early, affecting product quality. Thus, a more effective method of plastics removal should be implemented. In this approach, a proper treatment and method of recycling while some of the plastics are removed after the dismantling stage, prior to preheating to remove the electrolytes should be implemented. Removal of the entire plastics prior to preheating can lead to electrolyte leakage, hence, only a small fraction of the plastics should be removed, while the rest are removed during the pyrolysis stage. The plastics, PVDFs, if not removed properly may find their ways into the downstream products, thereby affecting the products purity/quality. This could explain why (or one of the reasons) products recovered by pyrometallurgy are of poor qualities.
- According to up-to-date literature, most researches and processes didn't account for the recovery of electrolytes, anodes (such as graphite), polymers, current collectors. Subsequent technological developments on recycling of LIBs should incorporate the recovery of these materials. Failure to recycle these materials and improper incineration have raised significant environmental concerns (such as toxic gas emission, water pollution, greenhouse gas emission). Thus, it's crucial to recycle or incinerate spent electrolytes, anodes, polymers and current collectors. Besides LIB manufacturing, recycled materials can find applications in other aspects of production.
- A green recycling method, namely a waste-for-waste approach, which is the most environmentally friendly scheme should be fully adopted. Lixiviant-containing food wastes (such as citrus fruits, apples, grapes, tamarinds) can be utilized in the leaching process of hydrometallurgical method to extract the valuable components of lithium-ion battery. Some researchers like [176], [179], [180] have implemented this approach to minimize energy consumption, carbon footprint and increase efficiency.

Table 7
A comprehensive summary of previous studies on direct recycling process.

Direct recycling process	Battery material	temp, ℃	time, hour	performances	References
Short annealing	NMC 111	800	4	Discharge capacity of 158.4 mAh/g at 1 C and 122.6 mAh/g after 100 cycles	[168]
Short annealing	NMC 523	850	4 h in O_2	Discharge capacity of 128.3 mAh/g after 100 cycles	
Hydrothermal	LCO	220	4	Retention capacity of 91.2 % after 100 cycles at 1 C (3–4.3 V); 141.9 mAh/g at 2 C and 130.3 mAh/g at 5 C	[169]
Hydrothermal	NMC 111	220	4	Discharge capacity of 158.4 mAh/g at 1 C and 122.6 mAh/g after 100 cycles	[168]
Solid phase heat treatment (under Ar/H_2 flow; Li_2CO_3 as lithium source)	LFP ($LiFePO_4$)	650	1	The first discharge capacity is 147.3 mAh/g (2.5–4.2 V); 140.4 mAh/g after 100 cycles at 0.2 C and capacity retention 95.32 %	[165]
Solid phase sintering (Li_2CO_3 as lithium source)	LCO	850		The first discharge capacity is 150.3 mAh/g (3.0–4.3 V) at 0.1 C; 140.1 mAh/g after 100 cycles	[170]
Hydrothermal	LCO	80	6	The first discharge capacity is 133.5 mAh/g; 99.5 % capacity retention after 40 cycles	[171]
Ultrasonic power (600w)	LCO			The first discharge capacity is 133.5 mAh/g; 99.5 % capacity retention after 40 cycles	

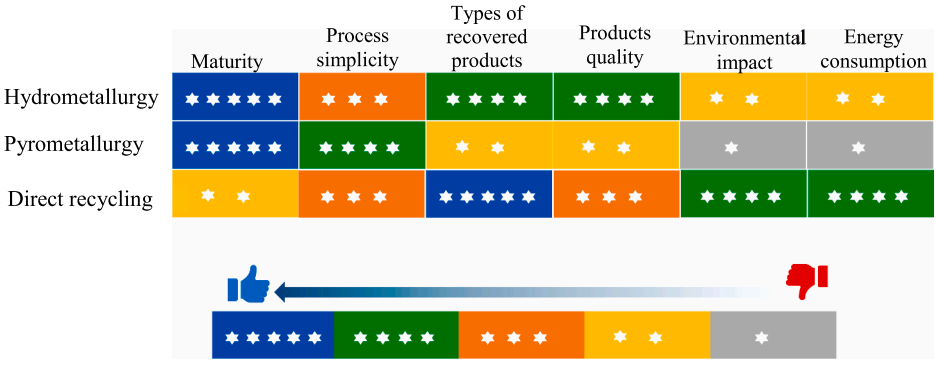


Fig. 8. Features of hydrometallurgy, pyrometallurgy, and direct recycling [176].

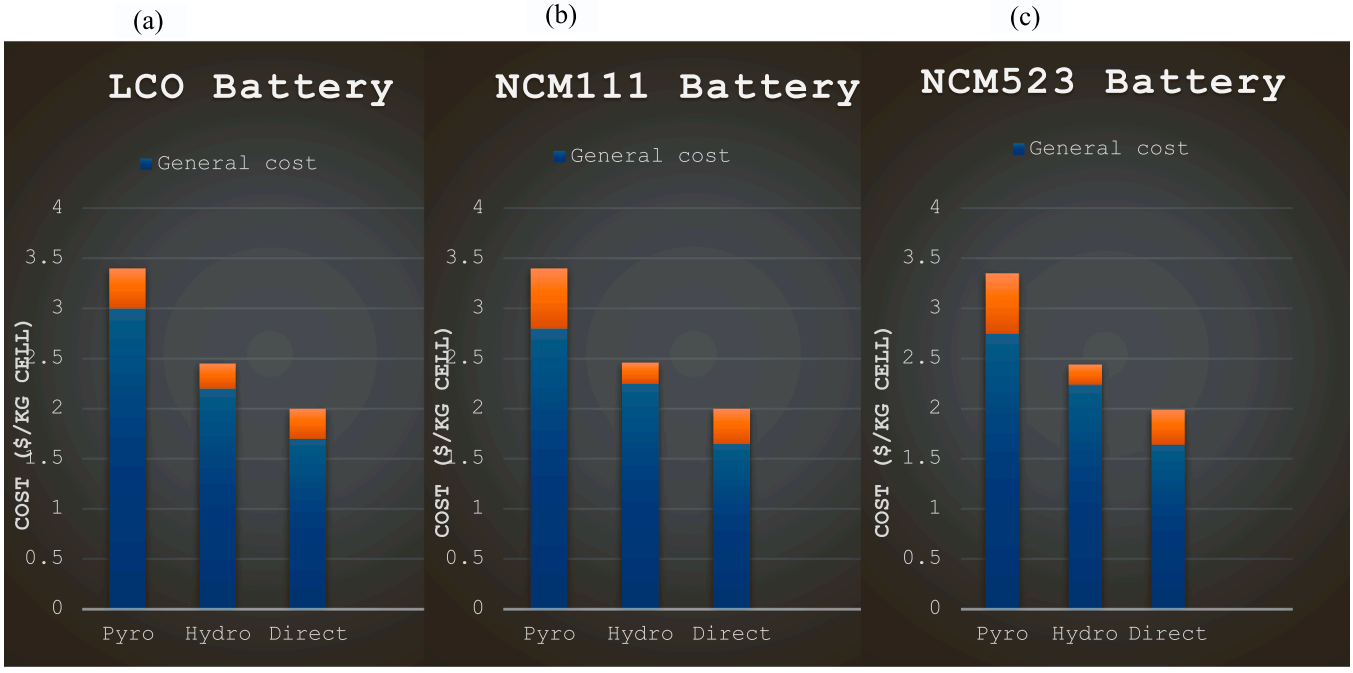


Fig. 9. Comparison of recycling costs of the three recycling methods for: (a) LCO battery (b) NCM111 battery (C) NCM523 battery [177].

5. Conclusion

Batteries (LIBs) are storage devices used to store electrical energy in the form of chemical energy. Expansion in the use of electronic devices and electric vehicles has increased the number of discarded LIBs at the end-of-life LIB. Various management options are available for spent LIBs, which include repurposing, remanufacturing, and recycling. Repurposing is employing a spent LIB to be used in another electrical

Table 8
Profit of the recycling methods (USD/kg cell) [177].

	Pyro	Hydro	Direct
LCO	8.2	9.3	14.9
NMC111	1.8	3.1	5.3
NCM 523	0.7	1.9	5.3

device which has a lesser energy demand than the initial purpose it was meant for. Remanufacturing is the restructuring of the battery pack and changing of damaged parts and cells of the LIB. Remanufactured LIB can serve its main purpose or other purposes. The ultimate fate of a spent LIB is recycling. The reason to recycle LIBs at the end-of-life spans from the need to conserve resources, to lives and environmental protection. The main recycling methods considered in this article are Direct, Hydrometallurgical, and Pyrometallurgical recycling processes. In direct recycling, the LIB active substances are recovered directly, while also maintaining the battery structure. Hydrometallurgy involves dissolving the valuable components of LIBs in an appropriate aqueous solution, and then reclaiming them by concentration and purification stages. While in Pyrometallurgy, the spent LIB is subjected to a very high temperature, then followed by separation processes. The direct method is only at the laboratory scale and still requires advancement. Hydrometallurgy and Pyrometallurgy are commercialized recycling processes. Pyrometallurgy is simple and reliable (in recovering Cu, Co, Ni, and traces of Fe), but has a very high emission rate. Hydrometallurgy has a very low emission rate and is super-effective. However, none of the three recycling processes is a perfect solution for recycling spent LIBs, with no emission rate, low cost, and low energy demand. Further research should be directed to address these.

CRediT authorship contribution statement

Henry C. Uzoma: Investigation. **Chukwuebuka E. Mgbemere:** Writing – review & editing, Investigation. **Daniel I. Udunwa:** Writing – review & editing. **Martin C. Etude:** Writing – review & editing, Writing – original draft, Formal analysis. **Alexander I. Ikeuba:** Writing – review & editing, Data curation, Conceptualization. **Chigoziri N. Njoku:** Writing – review & editing, Writing – original draft, Formal analysis. **Emmanuel Yakubu:** Investigation.

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

Data will be made available on request.

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