Review

Assessment of recycling methods and processes for lithium-ion batteries

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SUMMARY

This review discusses physical, chemical, and direct lithium-ion battery recycling methods to have an outlook on future recovery routes. Physical and chemical processes are employed to treat cathode active materials which are the greatest cost contributor in the production of lithium batteries. Direct recycling processes maintain the original chemical structure and process value of battery materials by recovering and reusing them directly. Mechanical separation is essential to liberate cathode materials that are concentrated in the finer size region. However, currently, the cathode active materials are being concentrated at a cut point that is considerably greater than the actual size found in spent batteries. Effective physical methods reduce the cost of subsequent chemical treatment and thereafter re-lithiation successfully reintroduces lithium into spent cathodes. Some of the current challenges are the difficulty in controlling impurities in recovered products and ensuring that the entire recycling process is more sustainable.

INTRODUCTION

In recent years, there has been rapid growth in the battery industry because of an increase in the demand of battery operated electronics, energy storage systems and more importantly, electric vehicles (EVs). This has resulted in the exponential consumption of lithium-ion batteries (LIBs) and in lithium becoming an essential raw material in various sectors. In the consumer market, secondary or rechargeable LIBs are widely used in manufacturing electronic devices including laptops, mobile phones, power banks, watches and lawn mowers. This is because of their relatively safe handling, high energy density, low self-discharge, and long life-cycles (Li et al., 2011b). Therefore, the development of technology has led to an increase in the use of LIBs, resulting in the need to recover Li, Co, Ni, and other precious metals. It is predicted that in 20 years the demand of lithium will become twice to six times the potential of its mineral deposits (Turcheniuk et al., 2018; Battistel et al., 2020).

Lithium batteries from consumer electronics contain anode and cathode material (Figure 1) and, as shown in Figure 2 (Chen et al., 2019), some of the main materials used to manufacture LIBs are lithium, graphite and cobalt in which their production is dominated by a few countries. More than 70% of the lithium used in batteries is from Australia and Chile whereas China controls >60% of the graphite production. Over 50% of the Co used is from the Democratic Republic of Congo (DRC). China imports the most unprocessed lithium minerals from Australia forming the largest bilateral trade pattern of unprocessed lithium (LaRocca, 2020). The global cobalt supply chain is also highly concentrated in the DRC for mining the cobalt and China for refining it (Fitch Solutions, 2021). Despite this, the global lithium consumption increased by 18% from 49,100 tons in 2019 to about 56,000 tons in 2020. By 2050, if the annual increase is maintained, lithium demand is predicted to be 63 million tons which will create approximately 50 million tons of lithium shortage as illustrated by Figure 3 (Bae and Kim, 2021; What's the big deal with lithium?; Lithium; Lithium industry worldwide; Lithium reserves worldwide 2010-2020). The increase in demand is expected to continue accelerating.

Moreover, there has been a global increase of waste from LIBs that contain an abundance of valuable metals which can be recovered for re-use. LIBs are lightweight in nature, which makes them an ideal energy storage component in electro-mobility applications (Susai et al., 2018). Basic recycling and unregulated disposal practices of e-waste frequently results in the production of toxic substances and the leaching of heavy metals into the environment. A large amount of LIBs from both EVs and electronics are still being

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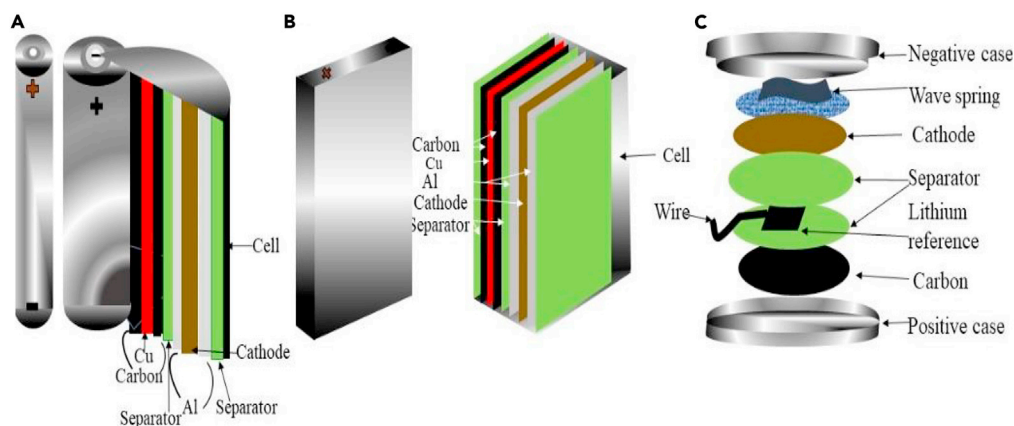
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Figure 1. Schematic picture showing the shape and components of various LIB configurations: A, cylindrical; B, flat; C, coin

disposed of in landfills such as in the case of Australia whereby 98% of LIBs were disposed on to landfills in 2012 and 2013 and only a small remainder was collected for recycling (King et al., 2018; King and Boxall, 2019). Taking into account the potentially damaging impact of LIBs and the abundance of valuable metals in them, LIB waste metal recovery is both environmentally and economically appealing (Erüst et al., 2013; Golmohammadzadeh et al., 2017; de Oliveira Demarco et al., 2019; He et al., 2019; Roshanfar et al., 2019; Yao et al., 2020). There are some commercial operations that are being employed to recover metals from LIB waste. However, although 99% of lead batteries are recycled, less than 5% of LIBs from EVs reach recycling plants and even less are recycled because they are more costly and difficult to recycle (Marchant, 2021).These battery packs are generally large and different manufactures use various battery assembling techniques.

Although major battery recycling companies are mostly located in Asia, Europe and North America, there has been a lack of international unified policies on battery recycling and use of recycled raw materials. This has been largely because of the rapid progression and innovation in the LIB market and an imbalance between new and mature markets. Furthermore, a global approach is not only politically challenging but also difficult to coordinate. However, further efforts to decarbonize transport have boosted domestic policies and regulations on battery recycling from EVs in certain regions, particularly the European Union (EU) and China. The EU enforced new regulations in January 2022 and China's aim to reduce CO_2 emissions per unit of GDP by 18% by 2025 and peak GHG emissions by 2030 (Khanna et al., 2021; Walstad, 2021) has also influenced a more urgent call for more batteries to be recycled with the increase of EVs. China strictly regulates the recycling supply chain and companies that comply with the regulations have a greater competitive advantage. Other regulations in China include new recovery rates for major battery metals. The recovery rate for nickel, cobalt and manganese must exceed 98% whereas the rate for lithium should not be below 85%. Rare earths are subject to a recovery rate of more than 97% (Changsha Sunda New Energy Technology Co. Ltd., 2019). The regulations are expected to increase purer supplies of battery metals.

The EU mostly relies on imported raw materials therefore their legislation affects the supply chain countries such as China. Its 12-member countries have put forward 2.9 billion euros in subsidies for the development of the EV battery industry in order to be less reliant on Asian imports (Petrequin, 2021). However, the battery recycling US policies are still lagging behind China and the EU. Hans Eric Melin of Circular Energy Storage Research and Consulting concluded that it is equally critical to ensure sustainability in the battery supply chain as it is to decarbonize the transport sector.

Given the steady increase of prices of precious metals such as cobalt and lithium and the correlation in demand of these metals from the E-bikes, portable devices and passenger EV industries, more effort is being placed into recovering these metals. This is to not only meet the future demand but also to tackle the financial aspect for these industries. However, end-of-life (EOL) batteries may take some time to reach the recycling stage and so primary resources will still be necessary in the immediate future The rise in demand is followed by a demand in EVs and plug-in hybrid electric vehicles (PHEVs) as presented in Figure 4.
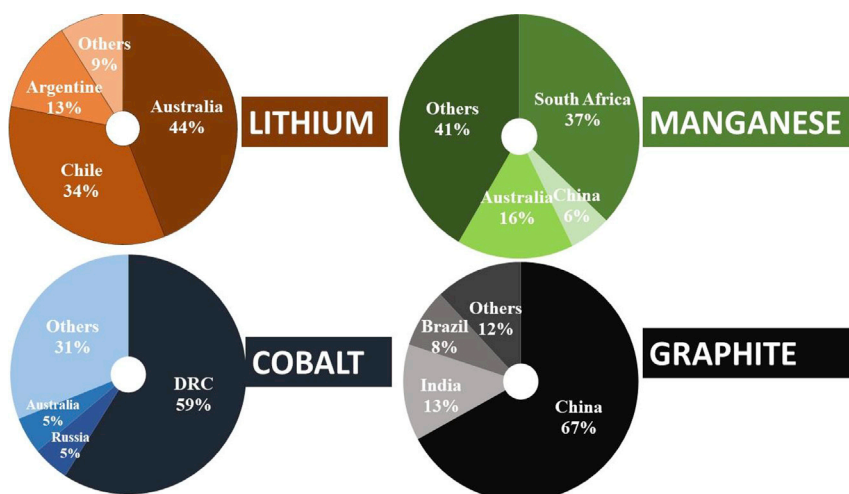
Figure 2. Global Production of main lithium-ion battery materials

The production of LIBs is expected to almost double in China by 2023 (Figure 4A) and the number of EVs is significant in the increase of overall lithium demand (Figure 4B). The increase in the output of LIBs is expected to continue; therefore, it has become crucial to research lithium recovery from EOL batteries. At present, it has already become evident that in comparison to the total lithium demand globally the increase in lithium-ion powered hybrid electric vehicles (HEVs) has been more drastic (Gruber et al., 2011). This is because of the market penetration of the vehicles and more environmental awareness, leading to a growth in the use of these vehicles (Eppstein et al., 2011).

In both China and the US, PEV market penetration was initially slow. However, since around 2014, China became the largest light-duty market for PEVs with 207,000 sales in 2015 as a result of aggressive policies and investment in this sector. By the end of 2017, cumulative sales reached over 1 million (Ou et al., 2019). This fast growing market in China has important implications for the global vehicle market and battery demand. The improvement of current recycling methods will guarantee that manufacturers spend less on metal purchases in the future by using the recycled material to make new batteries. The replacement time of lithium batteries has gradually been shortened because of the widespread use of LIBs which has resulted in a large number of discarded LIBs, especially in China (Zeng et al., 2012). In Beijing, 2.9 kilotons of lithium ion batteries were disposed of in 2013 and forecasts expect the number to increase at a rate of 26.7% (Jiang et al., 2015). In LIB studies, Co is ruminated as strategically important because of its military and industrial uses (Leon and Miller, 2020; Peeters et al., 2020). Co is the most expensive out of the four metals shown used in LIBs. It is about twice as expensive as Ni ($17,089 in Dec. 2020 (Bohlsen, 2020))and more than ten-fold the price of Al ($2,356 in Dec. 2020 (IndexBox, 2022)) with an average price of $31, 575 per ton in December 2020 as stated by the London Metal Exchange (London Metal Exchange, 2020) and was US$ 30,200 per ton in March 2014 (Barbieri et al., 2014).

Two aspects are involved in the impact of spent LIBs. Primarily, most cathode materials contain harmful organic electrolytes and hazardous heavy metals which are detrimental to the environment. Alternatively, spent LIBs contain valuable metals of which some of them such as lithium are more plentiful in LIBs than natural ores (Huang and Wang, 2019). Therefore recycling spent LIBs can alternately reduce pollution and preserve mineral resources.

Pyro- and hydrometallurgical methods are conventionally employed to recover metals from e-waste. Pyrometallurgy is economically feasible and conducive for large-scale operations. Most of the processes utilize high temperatures for metal recovery (Tuncuk et al., 2012; Chen et al., 2015; Yao et al., 2018; Ashiq et al., 2019). However, metals such as lithium, aluminum and manganese cannot be recovered at such high temperatures and the processes can also be energy intensive (Huang and Wang, 2019). In comparison, hydrometallurgical processes allow most of the metals contained within EOL batteries to be recovered after extraction and separation by leaching the metals into solvents. Studies show that strong inorganic acids for example, hydrochloric, sulfuric and nitric acids can be used to achieve 99% solubilization of lithium
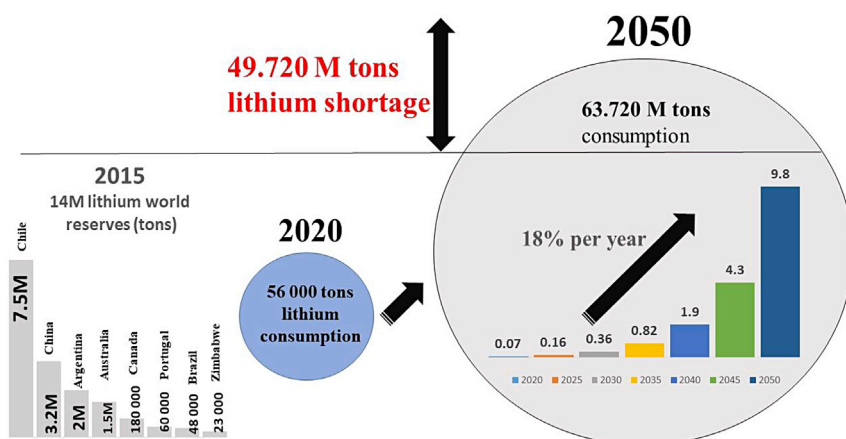
**Figure 3. Global lithium reserves and lithium depletion in 2020 and 2050**

and cobalt (Jha et al., 2013; Pagnanelli et al., 2016; Calvert et al., 2019). The process of recovering metals from metal-containing leachate solutions is quite complex. Other methods for metal recovery from LIBs have also been investigated such as membrane separation, precipitation, electrodeposition and solvent extraction (Li et al., 2019c). Using these methods, the combination of the recovery processes are energy consuming and require large amounts of reagents therefore, there has been increasing interest in further development of more environmentally benign recovery methods.

Numerous reviews have been published focusing on the technologies of conventional pyrometallurgical and hydrometallurgical processes (Wang and Wu, 2017; Zheng et al., 2018; Or et al., 2020; Bae and Kim, 2021; He et al., 2021). Some have reported on current developments of some processes (Makuza et al., 2021; Moazzam et al., 2021). Recently, few reviews have included both the physical and chemical processes as well as the environmental impacts of these processes (Huang et al., 2018; Fan et al., 2020; Zhou et al., 2020). However, a majority of these reviews focused on the separation and recovery process of positive electrode materials. Comprehensive reviews on the metal extraction and deep recovery of anode and cathode materials, including use of advanced technologies to reduce particle size to further liberate the electrode materials and environmental impact assessment during treatment and recovery of LIBs are lacking. This review article aims to address that gap.

CURRENT INDUSTRIAL RECYCLING METHODS

The most commonly employed industrial methods globally are the conventional pyro- and hydrometallurgical technologies which are combined or used separately (Klimko et al., 2020). The former is a process of thermal treatment employing high temperatures to cause physical and chemical transformations on spent LIBs for the recovery of metals (Makuza et al., 2021) whereby the latter is a chemical treatment method. Generally, a complete process of recycling includes sorting of the batteries, classification, mechanical treatment, followed by pyro-hydrometallurgical processes, waste disposal then recycling (Diekmann et al., 2017; Kuchhal and Sharma, 2019; Sommerfeld et al., 2020). Recycling companies normally cooperate with recycling organizations, for example, Call2recycle to guarantee sufficient recycling amounts. Some large-scale recyclers only produce a 'black mass' of active material that is then sold to other recyclers for pyro-hydrometallurgical recovery (Sommerville et al., 2021). The 'black mass' contains metal oxides and carbon. The carbon can be separated by froth flotation (Harper et al., 2019). The distribution and technologies used worldwide are presented in Figure 5 and it is apparent that more than half of the global battery recycling technologies are located in Europe and Asia.

As an example, a company called Umicore applies both pyro and hydrometallurgical processes to recycle spend LIBs by first feeding them into a smelter, skipping the pre-treatment process. Combustion occurs in the smelter and the gases produced are combated by advanced plasma technologies to prevent dioxin, furan and other pollutants from forming. Thereafter, high purity inert slags are produced and can be used as additives in construction e.g. in the steel, glass and ceramics industries. The principal products such as Co can be refined with hydrometallurgical procedures to produce lithium cobalt oxide which is a
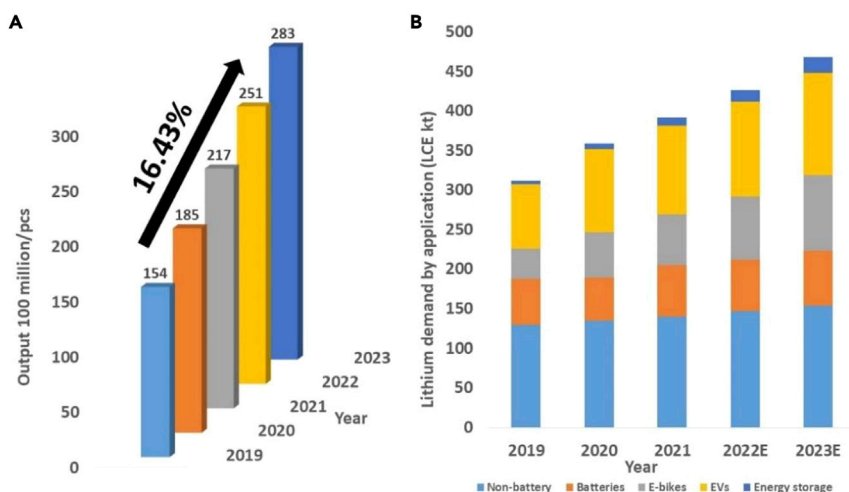
Figure 4. Production and demand of LIBs
(A) Output of LIBs in China (B) Lithium demand by end applications 2019-2023.

raw material for LIBs (Our recycling process | Cobalt & Specialty Materials, 2006; Umicore, 2021). Other companies such as Retriev Technologies and GEM use the methods shown in Table 1.

Retriev utilizes advanced technologies certified by the EPA for recycling all types of batteries and battery sizes. The spent LIBs are firstly discharged and disassembled then classified for recycling and scrapping. The company uses a patented cryogenic procedure whereby the batteries are chilled in liquid nitrogen to −200°C so that the lithium remains at a relatively inert state. Thereafter, the large pieces are crushed using hammer milling then subsequently placed in a caustic bath to neutralize the acidic components and dissolve the lithium salts. Lastly, lithium carbonate is produced as a raw material by use of salts and filter presses. The sludge produced is turned into lithium cobalt oxide (LCO) for new battery electrode materials (Retriev Technologies, 2021).

The spent LIBs at Accurec GmbH undergo a sorting process based on their chemistry into two different pathways. The lithium-ion automotive types of batteries are discharged until they contain less than 60V and their cooling systems are drained. Then, they are dismantled to modul/cell level before autothermal heat treatment. Through the other pathway, the lithium-consumer type manual batteries are also introduced to autothermal heat treatment after they are sorted into their subtypes and classified on the basis of potential thermal runway. The batteries with potential thermal runway undergo electrolytic discharge before autothermal heat treatment. The key features of the recycling techniques used at Accurec GmbH can be summarized into four main categories, highest safety at processing, no emission of environmental hazard electrolytes, batch wise processing of each lithium sub-chemistry and relatively low energy consumption (Accurec Recycling GmbH, 2021).

The LithoRec process is a battery recycling process that mainly aims to attain a high material recycling rate and focuses on energy efficiency. The main methods used are a combination of hydrometallurgical, mechanical and mild thermal treatment to regain almost all materials of a battery system (Diekmann et al., 2018). The batteries are first discharged by resistance then dismantled to expose the cells which are then shredded under inert atmosphere. The shredded material is immersed in dimethyl carbonate (DMC) to extract and recover the electrolytes before the iron parts are removed by magnetic separation. The remaining non-magnetic materials are separated into a section containing current collectors combined with active materials and another section of plastic foils mixed with separators. The binder is then burned off at temperatures between 400 and 600°C, the active materials and current collectors are separated by air jet sieves. Graphite is subsequently removed then lithium is leached out of the cathode material. An acidic mixture is used to dissolve the active material, which is then further refined by hydrometallurgical processes (Grützke et al., 2014).

As seen from the different recycling processes stated above, effective separation techniques decrease contamination of the active materials by other metals and improve the final recovery efficiency. Thermal
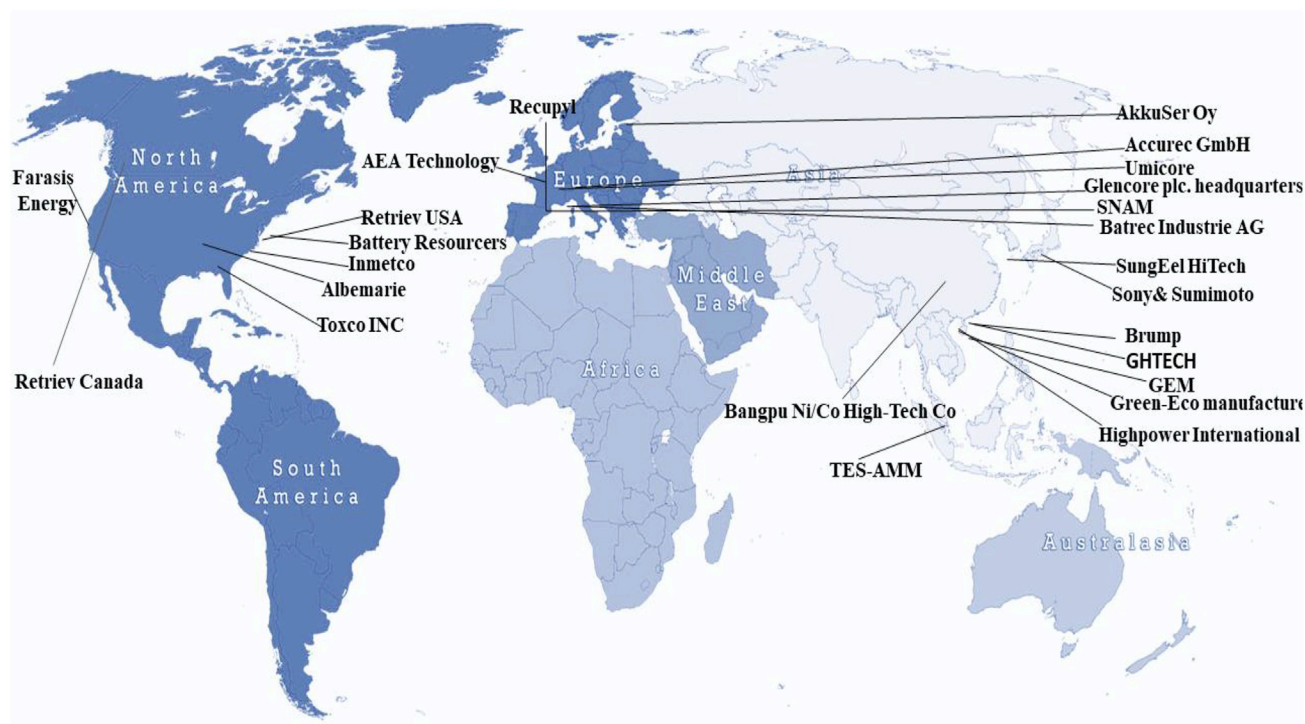
Figure 5. Distribution of spent LIBs recycling technologies

treatment, the dissolution process and vacuum pyrolysis are the most commonly employed. Several cathode and anode materials can be stripped from Al or Cu foil in the pre-treatment process. However, most of the material will still remain attached to the foil because of the $LiCoO_2$ electrodes in the LIBs containing $LiCoO_2$, an Al current collector and an electron-conducting carbon binder as well. Therefore secondary treatment is required and various alternatives have been experimented on and improvements have been made. These methods are divided into physical, chemical and direct recycling methods in this study.

PHYSICAL METHODS

For the recovery of lithium from spent lithium batteries, physical processes are usually applied to break up cathode materials from other components, e.g., current collectors and binders to facilitate recycling processes and reduce impurities (Saeki et al., 2004; Xu et al., 2008; Li et al., 2018). Common physical methods are organic solvent dissolution and mechanical separation based on several physical characteristics of LIBs such as magnetic properties, density and solubility (Dorella and Mansur, 2007; Li et al., 2018).

Mechanical separation

Mechanical separation is employed to remove outer plastics and shells from the spent LIBs with the aim of concentrating inner metallic components. The most widely used methods are magnetic separation (Wu et al., 2021), flotation (Zhan et al., 2018; Zhang et al., 2018a), crushing, shredding (Diekmann et al., 2017; Xiao et al., 2017a), sieving and gravity separation (Sun et al., 2021).

The crushing method presents potential hazards such as combustion or explosions thus inert atmospheres and low temperatures are applied as precautions for safety. Ordinarily, gases are released during crushing; Diekmann et al. (2017) found that EMC, CO_2 and DMC were the major gas components expelled during the crushing process. The battery health and the electrolyte component influenced the gas concentrations.

Moreover, crushing changes the surface properties of electrode materials in addition to changing the particle size and distribution of the battery components. This negatively influences the flotation separation process (Gratz et al., 2014; Yu et al., 2020; Zhang et al., 2021). During crushing, the electrolyte decomposes and the organic solvents, propylene carbonate and di-ethyl carbonate evaporate. An example is the

Table 1. Methods employed by recycling companies to treat spent LIBs

Conventional recycling methods	Company/PROCESS	Location	Resourcestype	Special Features
PYROMETALLURGY	Accurec GmbH	Mulheim, Germany	All (except Pb and Hg)	Vacuum thermal recycling
	AkkuSer Oy	Nivala, Finland	Metal powder	Two-phase crushing line
	Glencore plc. headquarters (former Xstrata)	Baar, Switzerland	Li and Ni	
	Inmetco	Pennsylvania, USA		
	Sony& Sumimoto	Tokyo, Japan	All	High-temperature calcination
	Umicore (VAL EAS™ process)	Brussels, Belgium	Li, Ni, Al	Single shaft furnace UHT technology
	SNAM	France	Li and Ni	Pyrolysis Distillation Recycling
	Toxco INC	Tennesse, USA	CoO	
HYROMETALLURGY	AEA Technology plc.	Oxfordshire, U.K	Li	
	Albemarie	North Carolina, USA		
	Bangpu Ni/CoHigh-Tech Co.	China	Cathode material	Leaching resynthesize method
	Batrec Industrie AG	Wimmis, Switzerland	Li and Hg	CO_2 protection
	Battery Resourcers	Massachusetts, USA		
	Brunp	Guangdong, China	Li and Ni	N/A
	GEM	Shenzhen, China	Li and Ni	N/A
	GHTECH	Guangdong, China		
	Green-Eco manufacture Hi-tech Co., Ltd	Shenzhen, China	Co and Ni	Leaching resynthesize method
	Highpower International	Shenzhen, China		
	LithoRec process	Germany?		High material recycling rate
	Recupyl	France, Poland, Italy, USA, Spain, Singapore	Li and Zn	Inert gas protection
	Retriev Canada	BC, Canada	All	Liquid N_2 protection
	Retriev USA	Ohio, USA	All	Liquid N_2 protection
	SungEel HiTech	Jeollabuk-do, South Korea		
	TES-AMM	Singapore		
Direct recycling	Onto process	Oregon, USA	Li and Zn	Ball milling
	Farasis Energy	California USA		

electrolyte, $LiPF_6$, which breaks down to form lithium fluoride and phosphor pentafluoride. The lithium contained in the lithium fluoride then dissolves in acid solution during leaching (Xu et al., 2008).

Shredding and milling have also been employed on LIBs whereby the batteries are shredded with a mill-cutter or shredder. A previous report has displayed that more than 50 wt % of $LiCoO_2$ is recovered in the >850 μm size region and contains a greater amount of Al and Cu (91.2 wt % and 89.7 wt %) which makes the feed not suitable for leaching. This milling method selectively liberates $LiCoO_2$ laminate which is strongly attached to its current collector compared to the graphite. It has also been discovered that the compression action of the cutting mill is partially responsible for $LiCoO_2$ remaining attached to the separator after liberation which then causes a reduction in the recovery rate of $LiCoO_2$ liberated in the smaller size fraction. Current collectors of new LIBs have better mechanical properties (Widijatmoko et al., 2020b) and the adhesiveness of their binders makes the finer size region less contaminated by Al and Cu compared toold LIBs.

Furthermore, both the positive as well as the negative electrode materials may be concentrated into the finer size region by wet and dry grinding without excessively crushing other components in the battery (Zhang et al., 2013; Chelgani et al., 2019). Size based separation can be done by selective liberation. The positive electrode active materials are concentrated by use of a suitable sieve size as a cut point. The impurities such as copper, iron and aluminum are concentrated above the cut point whereas the positive and negative electrode active materials are predominantly found below the cut point.

Numerous cut points were initially employed to concentrate positive electrode active materials, mostly between 250μm (He et al., 2017) and 2000μm (Li et al., 2009a). However, the 250μm cut point only recovered 56.38% $LiCoO_2$, From that study, the 250 μm cut point resulted in relatively low recovery of $LiCoO_2$ therefore for effective liberation by milling, the difference in morphological characteristics of new LIBs and spent LIBs were analyzed. It was observed that above the cut point 850μm, the graphite laminate was more readily liberated than the $LiCoO_2$ laminate which resulted in cleaner copper foils than aluminum foils. The morphological study was carried out using SEM and a classification of the liberated LIBs resulted in four categories as shown by Table 2. This was based on the size of active materials detached as well as the attachment of active materials on the current collector.

According to the categories, the bigger size fractions were classified into Category 1 and 2. These two categories also had the active materials laminate still attached whereas Categories 3 and 4 had detached active materials that remained attached by the binder. In the 850 μm-2360 μm size region, the positive electrode recovered was aluminum foil with $LiCoO_2$ laminate whereas the negative electrode found had minimum graphite lamination. Therefore, the size-based recovery rate demonstrates this by showing that the $LiCoO_2$ lamination to Al is stronger than the graphite lamination to the copper current collector. This results in the aluminum being concentrated in the larger size fraction as the lamination inhibits it from further breakage. However, the copper is concentrated in the smaller size fraction as shown. Evidently, copper has more suitable mechanical properties compared to aluminum but the $LiCoO_2$ lamination seems to improve the overall mechanical properties and prevents its breakage during milling. Alternatively, the cutting mill induced shear and tensile stresses which formed the nature of the Category 1 particles, and when the cutting mill induced stress-strain that did not dislodge the materials on the surface the particle size was reduced whereas the active materials remained resulting in Category 2 particles. In Table 2, the recovery rates of new and spent LIBs are compared. It is apparent that there is minimum contamination from Cu and Al in new LIBs in the <850 μm size region.

The lower contamination in this region is a result of the attachment of the polyvinylidene fluoride (PVDF) binder as well as the mechanical properties of the current collector. A great difference is in the >2360 μm size region for new and spent LIBs. The milled spent LIBs displayed a recovery rate of 59.8 wt % copper and the new LIBs displayed greater recovery of 73.6 wt % copper.

The cut point of 850μm was then employed in a different study where milling was utilized (Nie et al., 2015b). The LIBs were dried in an oven at 80°C to constant weight to remove electrolytes and moisture, followed by screening. Particle size distribution was conducted whereby the size fraction of <4750μm was selected and then the battery materials were calcined in a furnace. This fraction was then milled with a centrifugal mill with a 0.25mm grid followed by subsequent sieving whereby the cathode recovery size fraction was <850μm. The results indicate that 43.7% wt, 8.8% wt and 10.3% wt of $LiCoO_2$, Al and Co were recovered respectively (Widijatmoko et al., 2020a). The results still demonstrated low $LiCoO_2$ recovery thus, attrition scrubbing has been proposed as a second-stage liberation technique and is introduced in this paper as an advanced recovery technique.

During milling, either the occurrence of size reduction of the laminate happens while remaining attached to its current collector or the $LiCoO_2$ laminate is liberated as $LiCoO_2$-PVDF aggregates which are concentrated in the <850μm size region (Widijatmoko et al., 2020b). Conversely, the sizes of these aggregates are still larger than the actual $LiCoO_2$ particles in spent LIBs which are 1.50—7.80μm (Pavoni et al., 2018). Selective liberation is also dependent on the comminution technique that is employed (Hesse et al., 2017) whereby various techniques may result in differing size distributions. Selective liberation happens when the component breaks apart because of its physical and mechanical properties (Mariano et al., 2016).

Another preliminary mechanical method of separation of LIBs is sieving which is used in conjunction with milling. As an example, Al foils, Cu foils and plastics commonly exist in coarse fractions (>1 mm), whereby

Table 2. Characterization of classified LIBs powder

Category	Characteristics and Size	Size-based recovery rate
Category 1	Size reduction and detached active materials. The particles are in the size range of >2360 μm	
Category 2	Size reduction with active materials lamination, the particles are in the range of 2360–850μm	
Category 3	Active materials laminate that are held together by the binder and have been detached from the current collector. The aggregates for the particle size are in the range of 850–38 μm	
Category 4	Detached active materials laminate from the current collector with minimum aggregation for the particle size of <38 μm	

Source: Selective liberation in dry milled spent lithium-ion batteries (Widijatmoko et al., 2020b).

active electrode materials exist as fine fractions (<1 mm) (Wang et al., 2016a). Therefore, combining crushing and sieving as a mechanical pre-treatment process facilitates the separation of various components into a controllable range of particle sizes which benefits the acid leaching step according to Shin et al. (2005). In a study by Gaustad et al. (Wang et al., 2016b) five size fractions were investigated, mainly (<0.5 mm, 0.5–1 mm, 1–2.5 mm, 2.5–6 mm, and >6 mm). The effects of pre-sorting on the separation efficiency of spent lithium ion batteries were also studied. The results showed that Co was the only dominant element is the size fraction (<1 mm) with a portion of 85%. This portion is relatively high demonstrating that pre-sorting and sieving is highly beneficial to overall separation efficiency.

Flotation separation is a physicochemical separation process that works with the differences between the hydrophilic cathode materials and hydrophobic graphite anode materials. It separates materials based on hydrophobicity therefore the key is to control this parameter. During the bubbling process, the hydrophobic particles (usually graphite) are attached to the air bubbles and rise to form a froth layer that exits into a launder. The hydrophilic particles are left behind. Kerosene can be used to increase the hydrophobicity of graphite (Wakamatsu and Numata, 1991; Shin et al., 2020) and shows recovery rates of up to 90% of anode materials and 10–30% of cathode materials (Zhan et al., 2018). Conversely, there have been concerns about whether the original electrochemical reactivity of the recycled cathode materials is preserved upon exposure to moisture. Some reports have cited the benefits of water-based froth flotation on NMC cathodes. These studies show successful results without structural degradation (Kim et al., 2006; Xiong et al., 2013).

However, delithiation, surface species formation and lithium leaching has been known to occur (Zhang et al., 2011; Jung et al., 2017; Shkrob et al., 2017). Moreover, electrode materials are often coated with organic layers from the decomposing electrolytes after crushing (Zhang et al., 2014). This requires for the use of a reagent to regain the wettability difference, such as the Fenton's reagent which is effective in assisting flotation separation. Under an optimal condition of a Fe^{2+}/H_2O_2 ratio of 1:120 and a solid-to-liquid (S/L) ratio of 75:1, most organic layers can be removed (He et al., 2017). Another challenge is making this process more efficient while reducing the overall cost. Recently, an approach to tackle this issue has been proposed to recover pure coarse particles of Cu and Al (Saneie et al., 2022).

Froth flotation has also been employed as a direct recycling technique to separate blended cathode materials. Folayan et al. (2021) developed a froth flotation process to separate to separate ($LiNi_{0.333}Mn_{0.333}Co_{0.333}O_2$, NMC111) from lithium manganese oxide (LMO) materials. The results showed that at least 95% grade NMC111 and LMO were recovered.

Finally, gravity separation is used for components with different densities whereas the magnetic separation process eliminates magnetic impurities such as iron. However, in laboratory experiments, knives and saws are used to manually dismantle LIB cells. Magnetic separation is used for the removal of pieces of steel casing. A magnetic separator is used to eliminate aluminum foil pieces that remain adhered to the particles of $LiCoO_2$ (Or et al., 2020).

In summary, mechanical separation is employed before the metal leaching process to improve the recovery efficiency of target metals. It also eliminates the need to purify the leachate because the PVDF binder remains in the cake after filtration, it does not dissolve in acid solution (Xu et al., 2008).The main drawback of mechanical separation is that the components in the lithium batteries are all not separated completely because the batteries are composed of metals as well as organic and inorganic substances which penetrate into each other.

Dissolution

Dissolution is a simple method to separate cathode active materials from current collectors with binders. The most common binders are PVDF binders that are used for attaching the anode and cathode materials to current collector plates (Cu and Al foil) (Contestabile et al., 2001; Zeng and Li, 2014; He et al., 2015). These types of binders can easily be dissolved by organic solvents. Contestabile et al. (Contestabile et al., 2001) discovered that based on the dissolution process, the best separation method was the use of N-methyl-2-pyrrolidone (NMP) at 100°C for an hour. The anode and cathode materials were uncurled followed by treatment with NMP (Li et al., 2010a; Forte et al., 2020). The materials were then dried for 24hat 60°C to remove the PVDF binder and carbon from the cathode active material.

This is because NMP exhibits good solubility for PVDF (around 200gkg^{-1} of solvent) and has a high boiling point of about 200°C. Moreover, this permits for the recovery of Cu and Al in their metallic forms by filtration from the NMP solution. Generally, immersing electrodes in NMP is the most convenient way to separate active materials from Cu and Al. Thereafter, NMP can be easily vaporized.

A recent investigation by Li et al. (Li et al., 2009a) introduced ultrasonic washing as a method to improve the recovery rate of the dissolution process, particularly the recovery efficiency of Co while reducing pollution and energy consumption. Ultrasonic treatment should be conducted at temperatures below 55°C. In that study, the electrode material was immersed in NMP at 80°C for 1h then treated with ultrasonic technology for 20min. Further research has displayed that high-intensity ultrasonic delamination effectively blasts the active electrode powder from electrodes leaving the Al and Cu. That report showed that the lamination between the current collector and active layer could be broken within 0.5s. In addition, it was discovered that water-dispersible binders such as Styrene-Butadiene Rubber (SBR) were delaminated more rapidly (Lei et al., 2021).
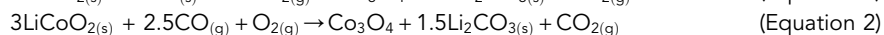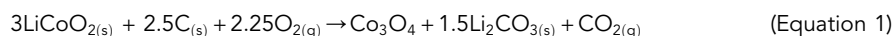
Ultrasonic technology can also be applied in chemical separation processes. NMP has proven to be an effective organic solvent for the dissolution of PVDF binders because they are both polar (Yao et al., 2018) but this single solvent cannot dissolve all kinds of binders. Although other organic solvents have been used to dissolve non-polar binders, organic solvents cannot remove all impurities and need subsequent calcining to burn off the remaining residues. However effective dissolution processes are in the separation of electrode materials from current collectors, they are also costly and the issue of highly toxic organic solvents may be addressed in future research.

Thermal treatment

Thermal treatment is another alternative method to pre-treat LIBs. There are two categories of thermal treatment, one that can be used for the removal of organic components, binders as well as electrolytes from spent LIBs. It is also employed to separate materials from current collectors. The other is pyrolysis which is applied to produce compounds or electrode materials (Li et al., 2016; Hu et al., 2017; Xiao et al., 2017a, 2017b) whereby the temperature and atmosphere of the thermal treatment can be controlled to obtain products with different purities and compositions depending on the nature of the components. Lee and Ree et al. (Lee and Rhee, 2002) modeled a two-step thermal treatment process. The LIBs were treated at 100–150°C for an hour to separate the materials from the current collector then calcination was performed at 500–900°C for 0.5–2 h to burn off binders and carbon. Thermal treatment is a simple operation technique that is suitable for large scale treatment but the combustion of carbon and binders produces toxic gases and smoke. Therefore, purification of gases is necessary to prevent pollution.

Vacuum pyrolysis is a type of thermal treatment used to separate electrode materials from binders. The organic materials evaporate to low molecular weight products weakening the adhesion between the Al foil and electrode materials, allowing for separation. Sun et al. investigated a vacuum pyrolysis method to completely peel off $LiCoO_2$ and CoO (cathode powders) from Al foils at 600°C then employed vacuum evaporation for 30min at a residual gas pressure of 1.0 kPa (Sun and Qiu, 2011). During pyrolysis, the volatile gases condense in the condensers while the non-condensable gases can be collected. Moreover, Xu et al. (Li et al., 2016) developed an in situ reduction pyrolysis method that calcined lithium cobalt oxide (LCO) and graphite in situ at 1000°C under N_2 atmosphere for 30 min. The products formed, Co, Li_2CO_3, and graphite were then separated by the wet magnetic separation method. In this study, thermodynamic analysis of the process verified the feasibility of the overall reaction therefore it has potential in industrial applications. This method can also be applied to recycle LMO and NCM cathodes (Xiao et al., 2017a, 2017b).

During thermal pre-treatment of LIBs, Li_2CO_3 is commonly formed by the following mechanism. During the combustion of graphite and organic compounds in the incineration process, high temperatures and the presence of oxygen result in the formation of CO_2 and CO. $LiCoO_2$ is reduced by carbon and carbon monoxide to Co_3O_4, Li_2CO_3, and CO_2 as illustrated by Equation 2 (Balachandran et al., 2021).

$$3LiCoO_{2(s)} + 2.5C_{(s)} + 2.25O_{2(g)} \rightarrow Co_3O_4 + 1.5Li_2CO_{3(s)} + CO_{2(g)} \qquad \text{(Equation 1)}$$
$$3LiCoO_{2(s)} + 2.5CO_{(g)} + O_{2(g)} \rightarrow Co_3O_4 + 1.5Li_2CO_{3(s)} + CO_{2(g)} \qquad \text{(Equation 2)}$$

On the other hand, pyrolysis takes place under nitrogen atmosphere and CO_2 and CO are formed by the carbothermic reduction of metal oxides (Jitka et al., 2012). Li_2O is formed during this reaction (Equations 3,
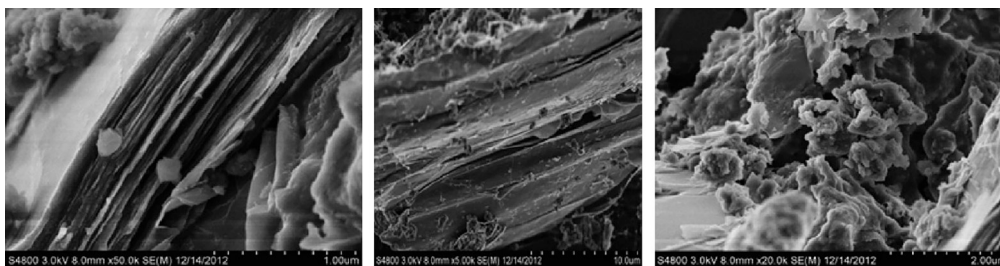
Figure 6. SEM images of the anode active materials of spent LIBs
Source: Leaching lithium from the anode electrode materials of spent lithium-ion batteries by hydrochloric acid (HCl)

4, 5, 6, and 7) which is then converted to Li_2CO_3 (Equations 8 and 9). Therefore, the main products are Li_2CO_3, Co and gases.

$$4LiCoO_{2(s)} + 3C_{(s)} \rightarrow 2Li_2O_{(s)} + 4Co_{(s)} + 3Co_{2(g)} \qquad \text{(Equation 3)}$$
$$4LiCoO_{2(s)} + C_{(s)} \rightarrow 2Li_2O_{(s)} + 4CoO_{(s)} + CO_{2(g)} \qquad \text{(Equation 4)}$$
$$2LiCoO_{2(s)} + 3C_{(s)} \rightarrow 2Li_2O_{(s)} + 2Co_{(s)} + 3CO_{(g)} \qquad \text{(Equation 5)}$$
$$2LiCoO_{2(s)} + C_{(s)} \rightarrow Li_2O_{(s)} + 2CoO_{(s)} + CO_{(g)} \qquad \text{(Equation 6)}$$
$$2LiCoO_{2(s)} + 3CO_{(G)} \rightarrow Li_2O_{(s)} + 2Co_{(s)} + 3CO_{2(g)} \qquad \text{(Equation 7)}$$
$$2LiCoO_{2(s)} + 3CO_{(G)} \rightarrow Li_2CO_{3(s)} + 2Co_{(s)} + 3CO_{2(g)} \qquad \text{(Equation 8)}$$
$$6LiCoO_{2(s)} + 5C_{(s)} \rightarrow 3Li_2CO_{3(S)} + 2CO_{2(G)} + CO_{(g)} \qquad \text{(Equation 9)}$$
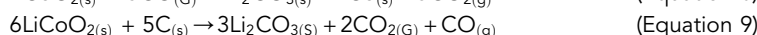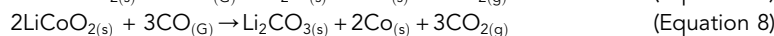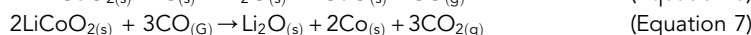
Pyrolysis is essential as a pre-treatment method because the high temperatures decompose organic material in the electrodes such as the binder, However, in some cases, the binder and organic ele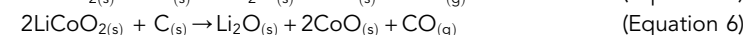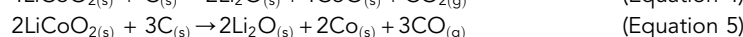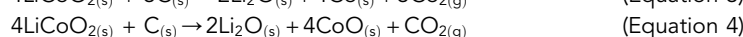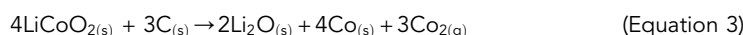ctrolytes are not completely removed and require further treatment as presented in Figure 6 (Guo et al., 2016), the morphology of graphite shows no change after the charge and discharge cycles. The particles are not smooth and have sticky substances. From the SEM results combined with the characteristics of typical anode material, it was deduced that the sticky substances contain PVDF binders and organic electrolytes.

PVDF binders have good mechanical performance and chemical stability, therefore are difficult to react with most strong organic acids. Efficient thermal treatment reduces issues during the solid-liquid separation step after leaching, and increases the recovery efficiency.

To validate the redox reaction and the change in structure of the electrode active materials caused by thermal treatment, the electrode active materials are characterized by XRD analysis, before and after treatment. Generally, the XRD spectra of active positive electrode materials before thermal treatment have a hexagonal lattice. However, after thermal treatment, the diffraction patterns of the electrode materials commonly contain only diffraction lines identified as metal oxides (Yang et al., 2016). This shows that the structures are destroyed during thermal treatment. For typical cathode materials, such as $LiCoO_2$, the carbon binder is burnt off by calcination above 700°C. Mechanical treatment combined with thermal treatment concentrates the electrode active materials in powder form and results in cost reduction and safety in shredding.

Thermal treatment of both positive and negative electrode materials in a high purity nitrogen environment considerably improves the recovery efficiency of valuable metals. The binder and the carbonaceous conductor can be completely burnt off by controlling the temperature between the melting point of current collectors and the thermal decomposition temperature of binders. Thermal treatment results in the separation of the current collectors, Al and Cu, from the active materials with minimum contamination and can be easily recycled after washing. It also changes the molecular structure of the positive electrode active materials and aids subsequent leaching by reducing the charges of transition metal ions in the positive electrode active materials.

Advanced methods

Attrition scrubbing is conventionally used in glass, mineral and water treatment industries to remove adhering sludge (Cole et al., 1981; Valchev et al., 2011) and has recently been introduced as a secondary liberation technique to enhance single-stage liberation performed with a cutting mill in lithium recovery from spent LIBs. Single-stage liberation has resulted in the recovery of 43.7 wt % $LiCoO_2$ in the size fraction

of <850 mm as well as 10.6 wt % and 9.0 wt % Cu and Al respectively in the size fraction >850mm (Widijat-moko et al., 2020a). The low recovery and lack of liberation is a result of the particles remaining adhered to the Al current collector therefore attrition scrubbing has been found to be an efficient solution.

In the application of attrition scrubbing, inert silica sand may be used as the abrasive to allow the $LiCoO_2$ to be concentrated in the finer size region then this step is followed by hydrometallurgical processes. A 56-U resistor can be utilized to discharge the batteries until the voltage reaches below 0.3V for safety reasons (Widijatmoko et al., 2020b). Thereafter the batteries are crushed with an 8mm grid crushing mill. The volatile organic materials in the batteries are removed by drying the samples in an oven at 80°C to constant weight then the dried sample is split into aliquots with a static rifle that has a chute size ranging from 31mm to 160mm. The representative sample obtained is screened for ferromagnetic materials with a cylindrical rare earth magnet attached to a PVC pipe.

Employing attrition scrubbing for 2.5 min enhances Co liberation toward Al by 36.6% and Cu by 42.6%. After 20min the $LiCoO_2$ recovery can be as high as 89.8 wt % with 9.0 wt % Al and 11.2 wt % Cu. The cut point is also considered in the recovery of lithium cobalt oxide. According to the morphological analysis of the particles and previous studies, a cut point of 850μm has been accepted. For attrition scrubbing, the $LiCoO_2$ particles remain laminated to the aluminum current collector above the cut point whereas they are in aggregate form below the cut point. Therefore, a smaller cut point (38μm) has been studied. Over 80 wt % $LiCoO_2$ particles with 7.0 wt % Al and 6.1 wt % Cu can be obtained in the <38μm size fraction therefore attrition scrubbing is a suitable second stage liberation technique (Sunil and Dhawan, 2019; Widijatmoko et al., 2020a; 2020b). Moreover, the recovered $LiCoO_2$ particles have minimum contamination from Cu and Al which reduces the need to purify the leachate during the hydrometallurgical process (Atia et al., 2019; Wang et al., 2019). The Cu and Al can be recovered by electrostatic separation (Kelly, 2003; Haldar, 2018). Attrition scrubbing is still being carried in a lab scale therefore further studies can focus on the scaling up of this technique and also testing it in a pilot scale.

An overview of studies done using the physical processes (Table 3) shows that mechanical separation plays an important role in improving overall recovery efficiencies. Recently, thermal treatment, especially vacuum pyrolysis has been favored over dissolution and advanced liberation methods show potential to improve recovery when used in conjunction with incineration and pyrolysis.

CHEMICAL METHODS

Chemical separation methods include leaching, thermal treatment and mechanochemical processes. These methods are then followed by purification. Purification comprises electrodeposition, chemical precipitation and solvent extraction to recover Co and Li compounds.

Mechanochemical reactions

These are chemical reactions generated by mechanical energy usually supplied in high-energy milling (Baláž et al., 2013). These reactions are usually a pre-treatment process which are used for the reduction of particle size and to break crystal structures of spent lithium ion batteries. This forms more stable compounds, facilitating the subsequent leaching process (Ou et al., 2015; Wang et al., 2016a, 2017b; Guan et al., 2017). The mechanochemical reaction method is advantageous with $LiFePO_4$ (LFP) cathodes which have a stable structure therefore are difficult to dissolve in acid. Yang et al. (2017) studied a mechanochemical (MC) reaction as a pre-treatment process of recycling spent LFP batteries. Disodium ethylene diamine tetra-acetic acid (EDTA-2Na) was selected as the grinding acid and the experimental parameters such as acid concentration, activation times, S/L ratios, mass ratios of cathode powder to EDTA-2Na, etc., were systematically investigated. The results showed that using MC reactions as a pre-treatment method improved the leaching efficiencies, for example, Li recovery efficiencies were enhanced from 60% to 94.29% and Fe from 40% to 97.67% with grinding time of 5h, mass ratio of LFP to EDTA-2Na = 3:1, and S/L ratio = $40gL^{-1}$. During the MC reaction, the crystal structure of the LFP cathodes was destroyed as shown by the XRD and Fourier transform infrared (FTIR) analyses in Figure 7.
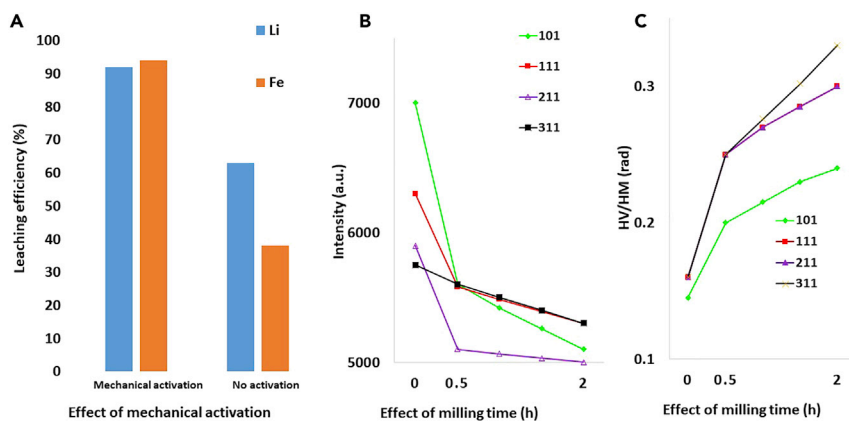
Leaching

LIBs are generally composed of leachable and non-leachable materials based on their solubility and ability to be deconstructed to their elemental forms during the leaching step. The current collectors, positive

Table 3. Summary of physical processes

Physical processes	Features		Results
Mechanical separation	Ball-milling	• Cathodes were ball-milled then sieved	(Golmohammadzadeh et al., 2017)
	Crushing & Planetary ball milling	• Size-reduction and sieving	(Hu et al., 2017)
	Milling & sieving	• Size fraction <850 µm	43.7 wt % of LiCoO$_2$, 8.8 wt % Al and 10.3 wt % Cu (Widijatmoko et al., 2020b)
	Shear crusher	• Al enriched component (>2 mm)	(Zhang et al., 2014)
		• Cu and Al enriched component (0.25–2 mm)	
		• Co and graphite enriched component (<0.25 mm)	
Dissolution	NMP	• Cathodes were treated with NMP at 90°C: 120min	(Chen et al., 2017)
		• Cathodes were treated with ultra-sonication in NMP at 100°C for 1 h	(Yang et al., 2016)
		• Cathodes were treated with ultra-sonication in NMP at 70°C, S/L ratio of 1:10 g/mL, ultrasonic power 240 W for 90 min	LiCoO$_2$ 98.99% (He et al., 2017)
	DMF	• Cathodes were treated with DMF under fierce stirring at 70°C; 120min	(Xu et al., 2014)
Incineration/Pyrolysis	Oven	• Cathodes were treated at 450–600°C; 60min	64.29%. Co$_3$O$_4$ (Barbieri et al., 2014)
		• Pyrolysis; 600°C;15 min	(Zheng et al., 2016)
		• 250–300°C for 30 min	Nickel 98%, Cobalt 99%, Manganese 84% (Yang et al., 2016) (Meshram et al., 2015)
	Pyrolysis		82.4% Li, 81.6% Co and 90.7% Cu (Sommerfeld et al., 2020) 96.7% Li, 81.67% Co and 67.38% Mn (Sunil and Dhawan, 2019)
Vacuum pyrolysis	30°C for 30 min		(Sun and Qiu, 2011)
Attrition scrubbing	20 min attrition Scrubbing		89.8 wt % LiCoO$_2$, 9.0 wt % Al, 11.2 wt % Cu (Widijatmoko et al., 2020a)
Triboelectric separation	One-pass electrostatic Separation		97.65 wt % metal (Cu and Al) (Widijatmoko et al., 2020a)

electrode materials and iron are leachable whereas polymeric materials and graphite are not leachable. Several studies (Chen et al., 2019; Roshanfar et al., 2019) have utilized manual dismantling to recover the positive electrode materials, which is practical for a laboratory scale recycling process but not suitable at an industrial level. Therefore it has become important to explore recovering positive electrode active material concentrate that is leachable without using manual dismantling. During leaching, the pH can be adjusted between 4.5 (Joo et al., 2016a) and 5.5 (Chen et al., 2011) with NaOH to remove impurities such as Al, Cu, and Fe. According to Sa et al., for a nickel, manganese cobalt (NMC) battery, the required impurities standard is about 5 wt % (Sa et al., 2015).

During the leaching process, carbon floats in acid solution and does not dissolve; therefore, it can be separated by filtration. For further refining, leaching is used in the transfer of metals from active materials to

Figure 7. Leaching efficiency of Fe and Li from different samples
Source: A Closed-Loop Process for Selective Metal Recovery from Spent Lithium Iron Phosphate Batteries through Mechanochemical Activation

solutions. Acid leaching is a standard procedure in a plethora of recovery processes and it can be selective toward specific elements. In the process of recycling spend LIBs, active materials such as Co and Li are transferred into solutions and alkaline leaching can be used to dissolve Al foils where the materials have not been removed from the current collectors beforehand (Gratz et al., 2014). Because of the formation of resultant stable metal complexes, leaching can be utilized to dissolve specific elements like Ni and Co (Ku et al., 2016; Zheng et al., 2017b; Chen et al., 2018; Qi et al., 2020). Leaching is considered as a chemical pre-treatment process and has significant influence on the overall recovery of metals.

Inorganic acid leaching

High leaching efficiencies of Li and Co have been achieved by inorganic acid leaching with H_3PO_4 (Belboom et al., 2015; Pinna et al., 2017; Jung et al., 2021), HCl (Li et al., 2009b; Guo et al., 2016; Liu and Zhang, 2016; Wang et al., 2019), HNO_3 (Lee and Rhee, 2003; Jung et al., 2021), and H_2SO_4 (Castillo et al., 2002; Shin et al., 2005; Zhu et al., 2012; Jha et al., 2013; Zheng et al., 2016). Generally, leaching with reducing agents such as glucose, H_2O_2, or $Na_2S_2O_3$ can increase the leaching efficiency by the conversion of metal oxidation states to more soluble states such as converting Co^{3+} to a more water soluble state, Co^{2+}. The equation for leaching with H_2SO_4 is as follows (Nan et al., 2005):

$$4LiCoO_2 + 6H_2SO_4 \rightarrow 2Li_2SO_4 + 4CoSO_4 + 6H_2O + O_2\uparrow \qquad \text{(Equation 10)}$$
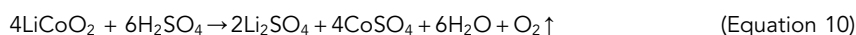
The leaching efficiency of cobalt increases with the H_2SO_4 concentration and reaction temperature therefore these need proper regulation. H_2SO_4 alone can leach up to 98% Co and Li at 70°C for 360min and with an S/L ratio of 1:5 (Nan et al., 2005). Further research has combined the use of H_2SO_4 with H_2O_2 (Sun and Qiu, 2011) and $N_2S_2O_3$ (Wang et al., 2012) to achieve the recovery of more than 99% Co and Li. A comparison between leaching efficiencies of sulfuric acid, nitric acid and hydrochloric acid revealed that H_2SO_4 had better leaching effect than HNO_3 (Sakultung et al., 2007). HCl can also be used to recover (Guo et al., 2016) lithium and cobalt from spent batteries but the reactions generate toxic gases like Cl_2 which pollute the environment.

Organic acid leaching

On the other hand, leaching with organic strong acids eliminates secondary pollution and harmful gas emissions without foregoing the high leaching efficiencies (Li et al., 2010b; Sun and Qiu, 2012; Nayaka et al., 2015; Zeng et al., 2015; Chen et al., 2016b; Gao et al., 2017; Zhang et al., 2018b). Organic substitutes are also degradable and do not require subsequent treatment. They also decrease the requirements for production equipment. Recently, citric acid, DL-malic acid and ascorbic acids are among the most commonly used. Citric acid with H_2O_2 can achieve a recovery rate of up to 99.8% Li and over 96% Co (Golmohammadzadeh et al., 2017).

Although citric acid is a weak acid, it's leaching effect is evident. Golmohammadzadeh et al. (Golmohammadzadeh et al, 2017) highlighted that the particle size distribution demonstrated that before leaching,

about 80% of the cathode particles were coarser than 10 μm and 90% of the particles were 55.90 μm. After leaching, 90% of the cathode particles were 53.10 μm. The size of the particles remained practically constant with mean diameters of 27.16 and 26.49 μm before and after leaching, respectively. The cathode materials had agglomerated morphology and irregular shapes which indicated that even after leaching with citric acid, the residues of PVDF floccules remained from previous treatment. This is also the case with DL-malic acid, it does not completely dissolve the binder and Co_3O_4 (Li et al., 2010a). Further optimizing the parameters causes better leaching efficiency as well as lower energy and chemical consumption. The best conditions are using 1.25M citric acid, 1.0 vol %. H_2O_2, S/L ratio of $20gL^{-1}$ and a leaching temperature of 90°C for 30min (Li et al., 2010b). Adjusting these parameters in a selective extraction method can reduce the leaching time by 10min (Punt et al., 2021).

Hydrogen peroxide is a suitable reducing agent as shown in Equation 11 and it accelerates the forward reaction between citric acid and the two metals in solution as shown in Equation 12 shows that Co is able to form chelate with organic acids. Therefore, these two reactions and the dissociation of organic acids at high temperatures prove the acceleration of the chelate formation reaction of Co with organic acids (Li et al., 2014).

$$H_2O_2 + 2H^+ + 2e^- \leftrightarrow 2H_2O \quad E^0 = +1.78 \ V \tag{Equation 11}$$
$$Co^{3+} + e^- \leftrightarrow Co^{2+} \quad E^0 = +1.80 \ V \tag{Equation 12}$$
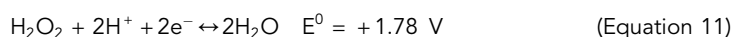
Li et al. (2012) discovered that ascorbic acid is able to act as both a reducing and leaching agent in Co recovery. In the experiments conducted, the optimum concentration of ascorbic acid was 1.25M, with a leaching time of 20min, temperature of 70°C and S/L ratio of $25gL^{-1}$. Under these experimental conditions, 98.5% Li and 94.8% Co were recovered in a short time under low temperature. This shows that using the leaching method in conjunction with organic acids is not only simpler, but environmentally friendly and utilizes less energy in comparison to conventional methods.

Water leaching

Water leaching has been recently employed to recover Li, Co and other precious metals from thermally treated LIBs. Water is used as a leaching agent with a low S/L ratio and at low temperatures. In this procedure, lithium is first recovered then the other metals remain in the filter residue because lithium salts show good solubility in water. During thermal treatment, carbon reduces the metal oxides to lower valence states which improves the leaching process (Balachandran et al., 2021). Water leaching reduces the need for additional reagents which minimizes the generation of secondary pollution. However, Liu et al., recovered Li_2CO_3 by using reduction roasting and hydrometallurgical recovery whereby the Li_2CO_3 was leached with water then H_2SO_4 leaching was used on the solid residue. 93.7% Li was recovered from a sample that was roasted for 30min at 650°C (Liu et al., 2018).

Thermal treatment in conjunction with water leaching can recover up to 99.5% lithium carbonate which contains 80% Li. Kuzuhara et al. recovered 90% Li_2CO_3 by incineration at 500-600°C (Kuzuhara et al., 2020). Another study showed that 82.7% lithium cobalt oxide was recovered after vacuum pyrolysis at 700°C for 30min and water leaching with an S/L ratio of 1:25 (Xiao et al., 2017b). Further research has been carried out to remove carbon in a recycling process whereby the cathode material undergoes thermal treatment for 5h at 500°C then it is leached with distilled water for 1h at 90°C (Paulino et al., 2008). 90% Li was recovered and the results showed that the removal of carbon enhances Li recovery which could be a strong adsorbent of Li^+ ions. The high recovery rates of Li by water leaching shows that the thermal pre-treatment process potentially simplifies subsequent hydrometallurgical processes.

Bioleaching

Bioleaching includes metallurgy, chemistry and biology. Recently, the bioleaching of spent LIBs has been widely attractive to researchers (Cerruti et al., 1998; Mishra et al., 2008; Xin et al., 2009; Horeh et al., 2016; Moazzam et al., 2021). Chemolithotropic and acidophilic bacteria are at the center of bioleaching whereby elemental sulfur and ferrous ions are used as energy sources for the production of metabolites such as ferric ions and sulfuric acid (Xin et al., 2009; Li et al., 2013) which can dissolve metals from spent secondary batteries. Moreover, fungi produce organic acids. Xin et al. (2009), has an exemplary study of this whereby Co and Li were mixed by a culture of sulfur and iron oxidizing bacteria. The results revealed that acid dissolution was the principal mechanism for Li bioleaching and that Fe^{2+} catalyzed reductions were also involved in the process.

Fungal bioleaching has proved to be effective in the recovery of precious metals from spent lithium batteries. Bioleaching using *Aspergillus niger* (*A. niger*) achieves a higher removal efficiency rate for heavy metals than chemical leaching. The growth characteristics of *A. niger* were investigated, including organic acid concentration, pH variation and biomass concentration. 100% Li was leached in the one-step and two-step-bioleaching processes whereas 95% was leached in spent medium experiments (Horeh et al., 2016). This demonstrates the potential of the hydrometallurgical route.

The use of bioleaching for large scale recovery of metallic components from e-waste such as Li could become more practical in the future. Despite the advantages of this process cited in literature (Borja et al., 2016; Bahaloo-Horeh et al., 2018), there is no sufficient research that show the industrial applications of bioleaching for waste resources recovery. At present, the complexity of e-waste composition (Baniasadi et al., 2019), low treatment capacity (Marra et al., 2018), gradual growth of microorganisms in waste (Wang et al., 2017b) and vulnerability to pollution (Zhang et al., 2018c) is restricting the industrial scaling up of bioleaching for metal extraction and recovery. Bioleaching is still being studied and expanded because of its possible role in pollution production and long periods of preserving bacterial cultures. In the long run, however, this process might be the most environmentally friendly.

Inorganic acid leaching is highly effective but the emission of secondary pollutants as well as the complexity of separation and purification steps is among its main drawbacks. On the other hand, organic acid leaching solves some of these issues but has other disadvantages such as the high cost of organic reagents and slower leaching processes. In comparison with these two leaching methods, water leaching is less cited in literature. This could be because it has been researched in conjunction with thermal treatment for effective results. Although some studies have considered bioleaching the most ecological leaching process, water leaching is also environmentally friendly and cost effective because it neither requires additional chemicals and reagents, nor is it an expensive procedure.

It is also important to consider leaching mechanism models to find the most suitable leaching route. The revised cubic rate law, the empirical model (Meshram et al., 2015; Pinna et al., 2017), Avrami model (Zhang et al., 2015a) and the shrinking core model (Jha et al., 2013; Chen et al., 2016b) are four leaching mechanism models that have been studied for kinetics. Mostly, the shrinking core model has been concluded as suitable for leaching analysis and researchers speculate that during the leaching process, particles go through a loosening-breaking-shrinking change. Moreover, residue layer diffusions, film mass transfers and surface chemical reactions can be used to control leaching reactions according to fitting results.

Chemical precipitation

Insoluble compounds are yielded from leaching solutions by chemical precipitation of precious metals. Generally, sodium carbonate, oxalic acid, ammonium oxalate and sodium hydroxide are the precipitants which are able to react with lithium and cobalt ions for the formation of insoluble precipitates of lithium carbonate (Zhu et al., 2012; Gao et al., 2017; Song and Zhao, 2018), lithium phosphate (Hu et al., 2017; Yang et al., 2018; Forte et al., 2020), cobalt oxalate (Sun and Qiu, 2012; Nayaka et al., 2015) and cobalt hydroxide (Pegoretti et al., 2017). Moreover, trace amounts of impurities such as aluminum or iron can be removed by chemical precipitation (Granata et al., 2012a; Joo et al., 2016b; Zheng et al., 2017a). This separation method relies on the different solubilities of compounds at specified temperatures and pH which require careful control during the process. An example of this is given by Sun and Qiu (2011) whereby in their study, a hydrometallurgical process was developed based on oxalate leaching, vacuum pyrolysis and precipitation. 98% $LiCoO_2$ was achieved whereby the optimum conditions were 1.0M oxalate solution at 80°C with an S/L ratio of $50gL^{-1}$. The mixture of active materials were leached for 120min then the Co was precipitated from $LiCoO_2$ to CoO directly as $CoC_2O_4 \cdot 2H_2O$.

Combining precipitation and leaching has proven to be a suitable method for valuable metal recovery. The combination of leaching and precipitation is a simple and adequate method to recover valuable metals. Wang et al. (Wang et al., 2009) investigated the separation and recovery of metals such as Ni, Mn, Co and Li from cathode active materials of lithium ion batteries. A $KMnO_4$ reagent was used to recover Mn as MnO_2 and $Mn(OH)_2$ then selective precipitation of $Co(OH)_2$ was conducted by adding 1M NaOH solution to reach an alkaline pH of 11. The remaining Li_2CO_3 aqueous solution was then precipitated by adding

saturated Na_2CO_3. The results showed that 96.294 wt %, 96.97 wt %, 97.43 wt % and 98.23 wt % of Co, Li, Ni and Mn were recovered respectively.

Chemical precipitation has wide applications because of its low cost, low equipment requirements and simple operation. In terms of recycling, selecting suitable precipitants and determining optimal conditions is crucial to avoid precipitate dissolution.

Solvent extraction

Solvent extraction is a two-phase system process whereby an organic and aqueous phase are introduced. Uneven distribution of the two phases aids the separation and after leaching, subsequent solvent extracts that are highly selective such as di-(2-ethylhexyl) phosphoric acid (D2EHPA) (Granata et al., 2012b; Chen et al., 2015; Vieceli et al., 2020) can separate selected transition metals from the leaching solution which allows the lithium to remain in the solution. Kang et al. (2010) combined extraction, acid leaching, and precipitation which resulted in 92% of Co recovery. First, Co was leached with 2M H_2SO_4 and 6vol % H_2O_2, then, through equilibrating with 50% saponified 0.4 M Cyanex 272, it was extracted from the purified aqueous phase at pH 6. Thereafter, the loaded organic phase with 2M H_2SO_4 was stripped and a solution of $96gL^{-1}$ Co was left behind. Lastly, evaporation/recrystallization was used to recover the pure cobalt sulfate.

This method can also be used to separate black mass from current collectors without corroding the current collector. Ethylene glycol has been investigated as a suitable solvent and recently, an ethylene glycol based solvent separation process has been performed (Bai et al., 2020a). The cathode and anode materials were recovered in several seconds and they remained structurally intact without any residues. The solvent could be directly reused creating a closed loop, cost-effective separation process.

The advantages of solvent extraction are that this process can efficiently produce metals with high purity and that it is usually performed at room temperature in a short period of time. On the other hand, the disadvantages are that the operations are complex and the solvents are costly. Therefore, developing cheap solvents and the cyclic usage of solvents will be a hot topic in future research. Apart from the main chemical processes (Lupi et al., 2005; Kang et al., 2010; Barbieri et al., 2014), other methods such as recrystallization and electrodeposition have been thoroughly investigated. Practically, to achieve higher recovery efficiencies, different chemical processes are usually combined together.

Electrochemical methods

Electrochemical methods have become an option for recycling LIBs because batteries contain suitable amounts of electrolytes. Electrochemical junction transfer has been employed in which Li^+ ions are selectively extracted from battery leachates by a porous material coated with an active intercalation $LiMn_2O_4$ matrix. Lithium intercalation uses an LMO junction to release Li ions on the other side of the junction into the recovery electrolyte resulting in a 90% transfer yield at $2.5mAcm^{-2}$ after 6h (Guyot et al., 2014). For the recovery of cobalt ions, electroreduction can be used to transform cobalt ions extracted from $LiCoO_2$ into cobalt hydroxide on a titanium electrode. A dehydration procedure is then used to obtain cobalt oxide (Myoung et al., 2002).

Electrochemical deposition technology can be used to regenerate $LiCoO_2$ and has several advantages such as relatively low cost, scalability and simplicity. In this procedure, LiOH was added as an electrolyte to a nitric acid leaching solution. A 2 mm layer of 0.5 µm $LiCoO_2$ particles was electrodeposited on a nickel substrate at current density of $1mAcm^{-2}$ for 20 h (Li et al., 2011a).

Overall, among the chemical processes, leaching has the highest efficiency of Li recovery as illustrated by Table 4 and thermal treatment has the highest recovery rate of Co. This shows that the combination of these processes could be essential to recover both lithium and cobalt used in spent LIBs.

DIRECT RECYCLING

Direct recycling has emerged as a method to maintain the original chemical structure and process value of battery materials by recovering and reusing them directly. Spent cathodes likely have low electrochemical performance because of lithium loss. Lithium loss could be caused by electrolyte interface layer formation,

Table 4. Summary of chemical methods

Chemical processes	Methods/Reagents	Features	Results
Solvent extraction	Mextral®272P	• Equilibrium pH of 4.5, 20 vol % Mextral®272P; 300s; A:O = 1:1	Co: 97.8% (Chen et al., 2015)
	Cyanex 272	• 50% saponified 0.4 M Cyanex 272; pH 5.5–6.0, A/O ratio =½	95–98% Co extraction ~1% Ni extraction (Kang et al., 2010)
	D2EHPA	• 70–75% saponification rate; 20 vol % D2EHPA;pH = 5,A/O ratio 0.5	Mn: 97% (Yao et al., 2018)
Chemical precipitation	$(NH_4)_2C_2O_4$ and Na_2CO_3	• Initial pH 2; 50°C; 60min	94.7% of Co and 71.0% Li (Zhu et al., 2012)
	Na_2CO_3	• pH adjusted to 2	Fe and Li (Zheng et al., 2016)
	Multi-step precipitation process	• 0.2 M dimethylglyoxime (DMG) solution, 0.5 M oxalic acid and 0.5 M phosphoric acid; 0.5 M $KMnO_4$	99%, 91%, 92% and 94% for Li, Ni, Co and Mn (Chen et al., 2016b)
Electrochemical Deposition	The initial charge and discharge capacity= 130.8 and 127.2 mAh g^{-1}		First cycle charge efficiency of 97.2% After 30 cycles, charge efficiency of 99.1% (Li et al., 2011b)
	Deposited Co^{2+} as $Co(OH)_2$ on a titanium electrode plate; constant potential; 200°C		Co_3O_4.(Myoung et al., 2002)

Table 4. Continued

Chemical processes	Methods/Reagents	Features	Results
Leaching	Organic acid leaching	• Citric acid (1.25M); 1% H_2O_2; 90°C; 30 min; S/L ratio 1:20	99% Li and 91% Co (Li et al., 2010a)
		• Tartaric acid (0.4M) and Ascorbic acid (0.02M); 80°C; 300min	>95% dissolution of Li and Co ((Nayaka et al., 2016))
		• Citric acid (2M); 1.25% H_2O_2; 60°C; 300 min; S/L ratio 1:30	99.8% Li and 96.46% Co (Golmohammadzadeh et al., 2017)
		• Iminodiacetic acid (IDA) and Maleic acid (MA); 80°C; 360min	>90–95% dissolution of Li and Co in the first 1h (Nayaka et al., 2015)
		• Ascorbic acid (1.25M); 70°C; 20 min; S/L ratio 1:25	98.5% Li and 94.8% Co (Li et al., 2012)
		• Citric acid (2M); 2% H_2O_2; 80°C; 90 min; S/L ratio 1:30	99% Li and 95% Co (Chen and Zhou, 2014)
		• Oxalic acid (1M); 1.25% H_2O_2; 95°C; 150 min; S/L ratio 1:15	98% Li and 97% Co (Zeng et al., 2015)
		• DL-malic acid (1.5M); 2% H_2O_2; 90°C; 40 min; S/L ratio 1:20	94% Li and 93% Co (Li et al., 2010b)
	Inorganic acid leaching	• HCl (4.0M); 80°C; 120min	97% Li and 99% Co (Li et al., 2009a, 2009b)
		• H_3PO_4 (7.0M) +4 vol % H_2O_2; 40°C; 60min;50gL^{-1}	99.9% Li and 99.7% Co (Chen et al., 2017)
		• H_2SO_4 (2.0M); 5% H_2O_2; 80°C; 60 min; S/L ratio 1:50	99% Li and 99% Co (Sun and Qiu, 2011)
		• H_2SO_4 (2.0M) +4.0% H_2O_2; 70°C.; 120min; S/L ratio 1/10	97.8% Mn, 99.4% Ni, 99.6% Co, and 98.8% Li (Nayl et al., 2017)
		• H_2SO_4 (2.5M); 60°C; 240min;100gL^{-1}	97% Li and 98% Co (Zheng et al., 2016)
		• H_2SO_4 (3M) + 0.4 g/g sucrose; 95°C; 120min;25gL^{-1}	100% Li and 96% Co (Chen et al., 2018)
		• H_2SO_4 (3M) + $Na_2S_2O_3$ (0.25M), 90°C; 180minS/L ratio 1:15	99.95% Co and 99.71% Li (Wang et al., 2012)
		• Cu-Fe^{2+}/Fe^{3+}-H_2SO_4-H_2O: 30°C; 120min	92% Co (Porvali et al., 2020)
	Deep-eutectic solvent	Choline chloride–citric acid DES (2 : 1 M ratio), 40°C; 60min; S/L ratio 1:20	$LiCoO_2$ (Peeters et al., 2020)
	Bioleaching	*Rhodotorula mucilaginosa*; heterotrophic yeast: 40days	92% Li (Sedlakova-Kadukova et al., 2020)
		Aspergillus niger; heterotrophic fungus; 40days	77% Li (Sedlakova-Kadukova et al., 2020)
	Alkaline leaching	5 wt % NaOH solution; room temperature; 240min; S/L ratio 0.1 g mL−1	99.9% Al dissolved (Chen et al., 2011)
		NH_4OH (4M); 60°C; 60 min; S/L ratio	97.8% Al and 64.7% Cu (Porvali et al., 2020)

surface degradation and parasitic reactions from surface impurities (Lou et al., 2015; Li et al., 2019b). Direct recycling typically requires the improvement of the electrochemical performance of EOL cathodes through regeneration. In this step, the composition and crystal structure of the cathodes is restored.

These processes are more efficient than pyro-hydrometallurgical methods because pyro-hydrometallurgical methods yield metal salts that need to be regenerated to form battery materials. Direct recycling enables recycling companies to retain the value of original battery materials at a lower cost. Theoretically, all the components of EOL batteries can be directly recycled including graphite, separators, Al sheets and electrolytes. Both cathode and anode materials can be obtained, reconditioned and used again in the manufacturing supply chain without additional processing. In comparison, for example, if a spent NCM battery is to be recycled, usually single metals such as Ni, Co and Mn are recovered using pyro-hydrometallurgical processes then the metals are synthesized to make new NCM cathode materials but with direct recycling, the final product is the NCM cathode material. The overall recycling steps are fewer and less energy intensive.

Several direct recycling methods such as the use of eutectic mixtures focus more on the separation of cathodes from current collectors, leaving the cathode materials intact. Processes using eutectic mixtures of lithium compounds to separate and recover active materials have also emerged. Common inorganic lithium compounds ($LiCl$, $LiNO_3$, and $LiOH$) have been studied and the results indicate that the highest peel-off efficiency (98.3%) is attributed to a $LiOH$-$LiNO_3$ eutectic system. It can be used to peel off NMC and LMO cathodes as well as regenerate them (Ji et al., 2021) which leads to a one-step separation and regeneration process. This experiment was performed at 260°C with a Li salts-to-cathode mass ratio of 10:1. The cathode material also underwent minimal change and remained structurally sound.

On the other hand, regeneration of cathode materials has been at the center of the development of direct recycling methods. Re-lithiation methods have been utilized to reintroduce lithium into cathodes and have been more successful in restoring batteries to pristine condition. Electrochemical re-lithiation has been applied to fill the vacancies in the layered structure of spent cathode materials with lithium ions. The lithium resource includes Li metals, concentrated Li solutions or cathode sheets in pristine condition. The use of aqueous electrolytes and subsequent thermal treatment has also demonstrated potential to recover EOL cathode materials while retaining the original morphology (Yang et al., 2020) of cathodes. On NCM material, ambient pressure re-lithiation with a eutectic Li-ion molten-salt solution has been performed successfully (Shi et al., 2019).

The Re-Cell Center for advanced battery recycling (ReCell Center, 2022) has made great progress in developing and evaluation numerous re-lithiation methods, namely, electrochemical, thermal, ionothermal, redox mediator and hydrothermal methods. Their research shows that the re-lithiation of artificially aged commercial cathode materials were restored to more than 95% of their original capacities. From these methods, thermal and hydrothermal re-lithiation have been more extensively researched and are thus presented in this work in greater detail and in Table 5.

Thermal re-lithiation

Thermal methods such as solid state sintering have been used on LFP (Wu et al., 2021) and LCO battery materials to regenerate the performance of cathode materials. These methods treat the spent battery powder as the precursor and the battery powder is calcined with a Li source such as $LiOH$ or Li_2CO_3. They are similar to the synthesis procedure in cathode powder production and can extensively heal the active materials. High temperatures are used to incorporate lithium ions into available sites to make up for lithium loss in the cathode materials.

Li_2CO_3 is most commonly used and has been successfully applied to recover $LiCoO_2$ from spent LIBs at 900°C in which the physicochemical properties as well as the attenuation rate of capacity, discharge capacity, attenuation rate of plateau and plateau retention at 3.6 V met the commercial recycling requirements (Nie et al., 2015a). The discharge capacity and experimental conditions are listed in Table 5. In another study, Li_2CO_3 was also used to increase the ratio of Co^{3+} in the cathode material and up to 95.78% $LiCoO_2$ and Co_3O_4 was recovered with an initial discharge capacity of 150.3mAh g^{-1} and 140.1mAh g^{-1} after 100 cycles (Chen et al., 2016a).At 850°C, Li_2CO_3 was used for 12 h and the discharge capacity was

Table 5. Summary of main re-lithiation studies

Direct recycling technologies	Lithium resource	Experimental conditions	Initial discharge capacity mAh g^{-1}	Final discharge capacity mAh g^{-1}/ (Number of cycles)
Thermal re-lithiation	Li$_2$CO$_3$	900°C; 12 h	152.4	149.9/(80) (Nie et al., 2015a)
		850°C; 12h	152.1	135.4/(100) (Shi et al., 2018b)
		850°C; 10h	150.3	140.1/(100 cycles) (Chen et al., 2016a)
		850°C; 4h	149.3	134.6/(100) (Shi et al., 2019)
		850°C; 4h; sintered in O$_2$	153.3	125.4/(100) (Shi et al., 2018a)
		800°C; 10h	165	133.3/(100) (Meng et al., 2019)
		750°C; 6h	−35	−35/(10) (Wang and Whitacre, 2018)
		800°C	126	107/(50)
		950°C	140	114/(50) (Han et al., 2021)
	/	100mL Cyrene 100°C for 1h Calcined at 600°C for 2h	194.0 mAh/g	158.0/(40) (Bai et al., 2020a, 2020b)
Hydrothermal re-lithiation	0.1M LiOH	Use of aqueous pulsed discharged plasma	132.9	126.7/(50) (Zhu et al., 2016)
	0.1 M LiOH	180°C; 2h	111	98/(100) (Gao et al., 2020b)
	1M LiOH; 1.5 Li$_2$SO$_4$ at 220°C; 4h	800°C; 4h	148.2	135.1/(100) (Shi et al., 2018b)
	2M LiOH at 220°C	–	131.5	129/(20) (Zhang et al., 2015a)
	/	LFP doped with 5 wt % graphene	161.4	–94.9% retention (100) (Jiang et al., 2022)

152.1 mAh g-1 after 100 cycles (Shi et al., 2018b). In a different investigation with the use of Li_2CO_3, the Li to Co ratio was maintained at 1.00 and the mixture was calcined at 800°C then the regenerated $LiCoO_2$ was coated with nanosized Al_2O_3 particles to improve electrochemical performance (Gao et al., 2020b). The Li/Co ratio from cell to cell should be determined to facilitate the amount of Li_2CO_3 that is required in order to avoid deficient or excess amounts of the lithium salt. During the reaction, Co_3O_4 is released from the decomposition of $LiCoO_2$ which then reacts with the Li_2CO_3 to form $LiCoO_2$ with improved cycling stability. The melting point of Li_2CO_3 is 723°C (Navarrete et al., 2022), therefore for improved cycling stability, experiments that utilize Li_2CO_3 as a lithium resource need to be performed at temperatures higher than 723°C.

At a lower temperature of 600°C, $Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$ scraps were regenerated by a simple and sustainable direct thermal process. Direct calcination, solvent dissolution as well as solution dissolution were carried out and the results showed that the highest retention was 96.7% after 100 cycles at 0.2C (Zhang et al., 2016). At the same temperature, $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ (NMC622) cathode scraps were treated with 100mL of Cyrene solvent, filtered and then calcined at 600°C for 2 h. The results showed that the initial discharge capacity was 194.0 mAh g^{-1} and that the recovered NMC622 retained 91.87% of the reversible discharge capacity (Bai et al., 2020b). Although thermal direct recycling is the simplest direct recycling method, it is still a challenge to determine the amount of Li that needs to be supplemented.

Hydrothermal re-lithiation

Hydrothermal re-lithiation utilizes lithium hydroxide solution to reintroduce Li into the spent cathodes. Ultra-sonication and aqueous pulsed discharge plasma are some of the technologies that have been used to accelerate lithium ion transfer during hydrothermal re-lithiation. For example, the aqueous pulsed discharge plasma method was used to regenerate LCO. 0.1 M LiOH solution and aqueous pulsed discharge plasma was used for 30 min on the cathode material to regenerate their electrochemical performance. The recovered LCO had an initial charge capacity of 132.9 mAh g^{-1}, discharge capacity of 126.7 mAh g^{-1} and 97.2% of this discharge capacity was retained after 50 cycles (Zhu et al., 2016). However, in this case, the discharge capacity of the first cycle is less than 130 mAhg^{-1} which shows that the electrochemical performance was not entirely regenerated. In addition, hydrothermal re-lithiation was applied in conjunction with ultrasonic regeneration on $LiCoO_2$ cathodes and the results showed a retention rate of 98.1% after 20 cycles (Zhang et al., 2015b).

A two-step hydrothermal regeneration process was introduced. The recovered products had an initial discharge capacity of 135.1 mAh g^{-1} and 91.2% retention rate after 100 cycles (Shi et al., 2018a). 1M LiOH and 1.5M Li_2SO_4 were used and the solution was heated at 220°C for 4 h then thermally treated at 800°C for 4 h (Shi et al., 2018b). To further reduce cost and greenhouse gas emissions, 0.1 M LiOH and 3.9 M KOH were studied. This was then employed in another investigation whereby 0.1M LiOH solution was used in a one-step hydrothermal process without KOH at a temperature of 180°C for 12 h for the re-lithiation of LMO. The results showed that the capacity retention of the regenerated cathodes was about 1.4% higher than that of pristine cathodes (Gao et al., 2020a). Moreover, the concentration of LiOH is more important than the reaction time or temperature (Wang and Whitacre, 2018). These results also demonstrate that a one-step process is sufficient to restore high electrochemical performance to used cathodes.

Cathode healing has also been investigated. It is a hydrothermal reintroduction of lithium to EOL cathode material rather than a solid-state addition of lithium (Mathew et al., 1998). It is applied to restore the EOL cathode powder without the use of ICP or lithium quantification. Wang and Whitacre (2018) have stated that cathode healing is better than solid state sintering in regards to phase purity and electrochemical capacity of the recycled material (Ji et al., 2021). Spent batteries have 10%–15% Li inventory deficiencies and undergo structural changes (Sloop et al., 2020). Therefore, research has shown that the Li vacancy defects (Li_v) and the Fe occupation of the Li site (Fe_{Li}) in spent LIBs affect the performance of LFP cathodes (Islam et al., 2005). The Li_v defects cause the oxidation of Fe^{2+} to Fe^{3+} and result in partial migration of Fe^{2+} to the lithium site which blocks the Li^+ diffusion pathway. Although there is loss in the storage capacity, the morphology of the crystal structure typically remains unchanged. Cathode healing enables the regeneration of the lithium capacity/conductivity structure property relationships.
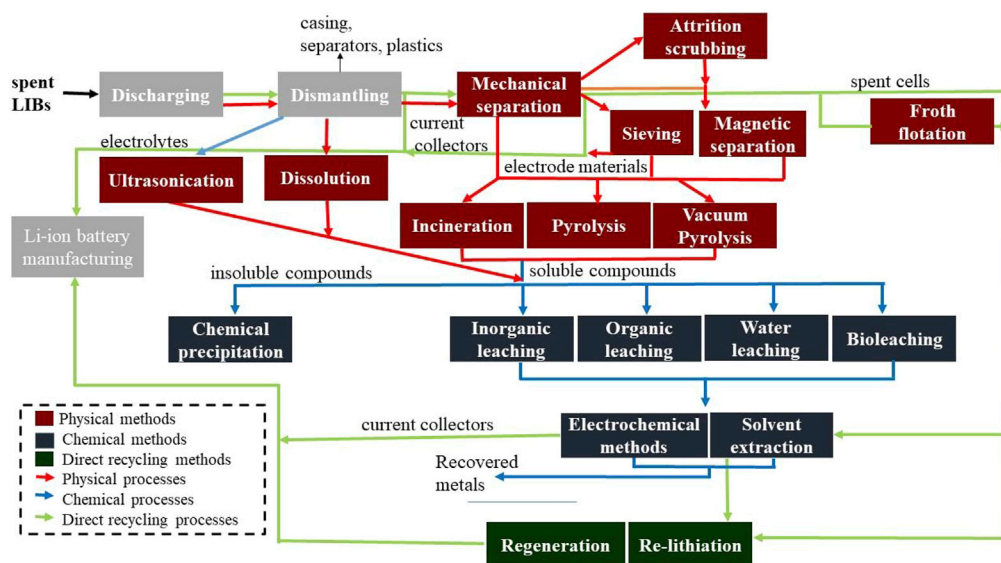
Figure 8. Recycling methods and processes flowchart

This method has been further up scaled to improve the cost and safety in a process that utilized aqueous methods to obtain the electrode material. The process includes first the extraction of electrolytes with CO_2 and shredding. Then, the plastics and other components are separated before froth flotation and finally the cathode material is heated and used to build new battery cells (Sloop et al., 2020). Cathode healing is also effective in directly regenerating $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ (Shi et al., 2018a).

However, the state of health of the batteries is crucial in direct recycling whereby low state of charge might not be advantageous. There are also challenges with different cathode formulations requiring tailored direct recycling processes to suit the metal oxide compositions in different batteries (Harper et al., 2019). However, direct recycling methods are more cost effective and favorable compared to pyro-hydro-metallurgical processes because they can efficiently regenerate EOL cathodes in less processes as displayed in Figure 8 and restore cathodes to comparable or better electrochemical performance than commercial pristine condition batteries.

ENVIRONMENTAL IMPACT EVALUATION

During treatment

LIBs from consumer electronics have an average service life of about 1–3 years or between 300 and 1000 (Wake up Roma, 2020a) charge/discharge cycles depending on the battery type and components. Li-ion EV batteries have a life span of between 5 years or 60 000miles and 8 years or 100,000 miles (EV Connect, 2021). Moreover, the average battery pack shelf life is 3–6 years (Wake up Roma, 2020b), therefore, they constantly require replacement which makes recycling crucial. However, the treatment of EOL batteries and recovery of precious metals pose potential health risks and environmental hazards. This is not only because of the original constituents of the batteries, but also new substances produced by side reactions through charge and discharge. The physical and chemical substances in the batteries eventually break down in the environment after the batteries have been discarded which leads to environmental degradation and pollution.

During thermal treatment, graphite carbon materials might react with strong oxidants which can generate gases such as CO and CO_2. Moreover, mixtures of dust and air might explode when exposed to heat (An, 2019). This is also the case if batteries are not sufficiently discharged and undergo mechanical treatment. This amount of excessive dust (Xiao et al., 2017b) causes dust pollution and an increase in the pH of the surrounding environment. Dust particles increase the pH of rain water (Wu et al., 2013; Hedin and Likens, 2017) because they contain bases which dissolve into cloud-water droplets. This in turn also affects the pH of surrounding soil.

During recovery

During the recovery process, different types of chemical reactants are added. Leaching agents are required such as sulfuric acid (Chen et al., 2015) and hydrochloric acid (Shuva and Kurny, 2013) as well as oxidative agents such as hydrogen peroxide (Huang et al., 2019). The production procedure of leaching agents and the treatment of excess reagents can be analyzed to alleviate the environmental burden of this process. In regards to acid production, taking phosphoric acid as an example, it was discovered that it has a highly radiotoxic element, uranium, that has the highest toxicity contribution of about 23%. Moreover, phosphoric acid waste contains barium which showed a 21% toxicity contribution (Belboom et al., 2015).

Life cycle impact assessment results for leaching LIBs with mild phosphoric acid showed that the climate change potential is less than 1kg CO_2-equivalent in terms of GWP_{100} which is relatively high but some processes result in a lower value when they are scaled up such as the thermal treatment process in which an oven has the capacity to dry many batteries instead of a small number of batteries when it is conducted in lab-scale. The end-point impact results are between 0.02 and 0.04 points which is lower than direct disposal (Sambamurthy et al., 2021).

As for sulfuric acid, it was observed that the treatment of batteries with sulfur has a negative impact on the environment, particularly on ozone depletion, freshwater eutrophication, human toxicity, ionizing radiation and land use as a result of the energy consumed in petroleum desulfurization (Mami et al., 2017). However, the majority of leaching agents transfer into metal salts thereby making the environmental impact more complex. Besides, the leaching agents, acids and alkaline solutions are added for neutralization in the recycling process as well as organic solvents for solvent extraction and metal salts for precipitation (An, 2019). However, the environmental impacts of these have not yet been studied extensively.

A study has shown that hydrometallurgical recycling has a much lower carbon footprint compared to pyrometallurgical methods (Sambamurthy et al., 2021). Another study indicates that incineration of plastics and electricity generation has the largest impact on GWP_{100} as well as HTP and TETP, respectively. However, plastics can be separated mechanically before incineration and the energy consumption can be reduced by employing more renewable sources of energy (Boyden et al., 2016).

In regards to energy consumption and its environmental impact on lithium battery recycling, calcination for cobalt recovery, crushing and grinding as well as leaching consume the most energy respectively. On the other hand, steps such as discharging and intermediate steps consume minimum energy and release less emissions.

DISCUSSION

Recently, technological developments have been made to improve battery recycling methods of spent LIBs, although the complexity of e-waste composition (Baniasadi et al., 2019), low treatment capacity (Marra et al., 2018), gradual growth of microorganisms in waste (Wang et al., 2017a) and vulnerability to pollution (Zhang et al., 2018a, 2018b) are limiting factors. Overall, chemical methods result in high recovery rates due to their selectivity and high purity of final products. They also use less energy while emitting less waste gas. However, they are usually long processes that produce more wastewater, therefore, the greatest challenges are optimizing the processes and wastewater treatment.

On the other hand, the advantages of physical methods are simple operation and short flow. They have no requirements for classifying the material into categories or limitations on the size of inputs. They also have shortcomings, such as the difficulty in recovering Li and Mn, high energy consumption and low recovery efficiency. Physical methods also produce waste gas which requires treatment and do not result in complete recovery, thus, need to be combined with chemical methods. Economically, they have high operational and equipment cost.

Most of the focus is on chemical methods such as chemical precipitation which are complex and challenging to scale up despite producing high purity products. Conversely, physical methods are favorable on an industrial scale as they offer simple operation despite the large amount of secondary pollution produced and higher energy consumption required. It is possible that more combinations of both of these processes will be developed for the production of higher purity final products from spent LIBs. Simpler and

economically feasible methods will be more favorable and more attention should be directed at secondary use of retired batteries. Ultimately, the future development of battery material and design will play an important role in the development of treatment technologies for spend LIBs.

Direct recycling and hydrometallurgical processes have the same major challenge, that is, efficiently separating cathode material adhered to Al foil as a result of the strong adhesion and chemical stability of the binders. Another issue is maintaining the morphology of the electrode material while avoiding impurities from side reaction products or current collectors. However, its advancements in high purity products, high recovery rates and low process consumption are driving forces in further developing direct recycling.

In the last decade, extensive research has gone into solving the issue recycling spent LIBs and vast progress has been made regarding the pre-treatment, extraction of precious metals and their recovery. However, several challenges and difficulties still remain and need to be addressed, such as:

(1) The dismantling and discharge procedures have to be handled carefully to avoid explosions. Procedures to safely dismantle battery packs, especially automatically haven't been fully explored. Impurities have a negative effect on the final products therefore it is necessary to control the amount and type of impurities in final products. Also the complexity and cost of methods that can achieve high purity products such as electrodeposition also needs equally efficient alternatives to be researched.

(2) Methods such as thermal treatment release toxic gases and waste solids which require further treatment.

(3) Currently, most studies concentrate on cathode materials and few deal with anodes and electrolytes because of the latter's high volatility. However, on the basis of environmental protection, these should be recycled and disposed of safely.

(4) Making recovery methods more economical is crucial as it incentivizes manufactures to recycle the spent secondary batteries.

High metal recovery from spent LIBs has been achieved using sustainable processes. Methods that have economic and environmental benefits have been developed by managing water and energy consumption. Over the years, studies have focused on improving the conventional recycling processes to make them safer, more eco-friendly and efficient to minimize the environmental impacts and maximize the economic aspects.

Another approach is investigating the redesign of LIBs that are being currently used. This might facilitate the disassembly process and the modifications may include features such as automated dismantling and smart separation. A study introduced a method to manufacture LIBs without hazardous organic solvents whereby the electrodes were fabricated using a water-based organic solvent (Li et al., 2020). Research on sustainable battery manufacturing is still in its early stages while automatic disassembly has been more widely discussed.

Automatic dismantling has been less favorable than manual dismantling because it results in the addition of impurities (namely, Al and Cu) in the subsequent leaching process (Golmohammadzadeh et al, 2017), but recently it has been explored further. Automated disassembly is being investigated for the liberation of electrode materials on an industrial scale to avoid contamination between electrode materials, current collectors and other battery substances. Because of the growth of EVs, manual disassembly will likely become less significant and consequently, potential for optimization in battery disassembly has been cited in several studies (Li et al., 2019a; Markowski et al., 2014; Wegener et al., 2015; Kay et al., 2019; Glöser-Chahoud et al., 2021). Automatic battery disassembly makes recycling of batteries potentially safer and more economically viable. Li et al. (Li et al., 2019a) has proposed an automatic disassembly system as a large-scale mechanical pre-treatment process for dismantling and separating battery materials in order to achieve higher material recovery with higher purity. The trimming module containing a blade set, trimming set and conveyor roller set. This module cuts of the front edge of the battery pouch and separates the electrode layers from the current collectors before further treatment.

There are still several drawbacks of automated assembly such as the lack of a unified standard for EV battery design. This brings about difficulties in regulating the automated process and optimizing it to accommodate different batteries with different structures and dimensions. Similarly, it is challenging for batteries to be properly classified for large-scale automated disassembly (Zhou et al., 2021). Battery designs differ from manufacturer to manufacturer. Besides, the technology required for this process has to be advanced and efficient. The systems have to be complex enough to tackle disassembly of batteries on a system, module and cell level. This might include the incorporation of a bit changing mechanism and camera-based detection of screws (Wegener et al., 2015). For this reason, hybrid human-robot workstations can be considered.

Alternatively, batteries could be remanufactured with segregated parts and separated electrode materials to ensure easier classification of the components before treatment. Then again, battery manufacturers are moving toward reducing cobalt amounts in cathodes. This might reduce the incentive for using pyro-hydrometallurgical processes, leaving direct recycling as the most viable recycling method (Gaines, 2017). Some methods have proven to be feasible in lab-scale but not pilot-scale therefore it is crucial to identify methods that provide industrialists with systems that have a production capacity that can cope with the future demand for recycling waste LIBs.

Despite the progress made in upscaling conventional methods, there are still very few medium to large-scale recycling facilities. This is mostly because of these five factors:

(1) Time-factor: Infrastructure development will take time as well as the processes to recycle and large numbers of batteries and their diverse components. On the other hand, recycling companies can take advantage of the extensive research that has already been done and of emerging or refined recycling methods to build effective recycling models.

(2) Economic factor: The construction of a typical recycling plant has been estimated to exceed US$3 million including the land requirement, machinery, utilities and labor (Estimated Cost to Build and Open a Recycling Plant in 2022, 2022) depending on the location and the size of the recycling plant. Taking into consideration, the cost of energy to run an e-waste recycling plant, cost of reagents or subsequent wastewater treatment methods, processing methods and so on, makes battery recycling an expensive industry, especially for a large-scale recycling facility. Moreover, it is cheaper to mine lithium than to recycle it, therefore another disadvantage lies in lack of resale value of low-purity recycled battery components especially lithium. Although there is some reasonable economic return from recycling batteries made with metals such as Co because of their market value, some LIBs such as LFP are made with relatively inexpensive metals therefore the economic value of their elements does not compensate for the high cost of recycling them.

(3) Technological factor: Some processes have been successful in lab-scale but are not yet viable for large-scale recycling such as manual dismantling which would be too time-consuming or multi-step processes that are too complex to perform on a large number of batteries. However, as more research is being done, not only on the main processes, but also on dismantling, discharging and so on, this might not be a great challenge in the future.

(4) Environmental factor: The production of toxic waste gas during thermal treatment causes significant air pollution when performed on an even larger scale. Although some chemical reagents can be reused directly, more batteries being treated means more reagents that would require subsequent wastewater treatment. Moreover, in some cases, heavy metal contamination has been detected in the soil, air and water near e-waste recycling sites (Li et al., 2015; Awasthi et al., 2016). In addition, the majority of industrial recycling companies focus on high value metal extraction than closed-loop recycling which highlights an environmental gap (Sommerville et al., 2021). The current circular economy of batteries should be further developed to encompass more green recycling methods.

(5) Safety factor: There are still concerns about the limitation of hazards such as explosions during transportation, sorting, processing and storage of a large number of batteries with different discharging requirements.

Currently, only a minimal number of recycling companies such as Retrieve and Umicore exist worldwide compared to the number of batteries being produced and future predictions of the number of batteries

that will require recycling. Because of policies and more interest in recovering precious metals from e-waste, new approaches are set to drive the large-scale battery recycling trend and more recycling companies have already risen to the challenge. Stena recycling (Stena Recycling to launch large scale battery recycling in 2023 - electrive.com, 2022), for example, has started construction on a new large scale battery recycling plant in Sweden that is set to be completed by 2023. Battery Resourcers also plans to open a $43 million commercial-scale recycling facility in August 2022 (McNees, 2022).

Conclusions

Scaling-up mechanical separation before recovery of precious metals from spent LIBs has several benefits such as the decrease in reagent consumption, reducing the amount of waste and improving the efficiency of the desired metals. Overall, recovery of metals from LIBs includes not only treatment but also source and processing control therefore with the increase of environmental awareness and technology, there more concentration of making green and simpler methods. In the future, selecting newly developed, anode, cathode and electrolyte materials as well as new battery designs. Therefore, the focus should be on the development of abundant and yet non-toxic materials for the production of LIBs so as to cause less environmental problems and will also facilitate better treatment of LIBs.

Limitations of the study

This review has mainly studied the key physical, chemical and two direct recycling methods of recycling spent LIBs. Other direct recycling methods such as cathode-to-cathode and ionothermal re-lithiation have not been included in great detail because of few supporting publications on these methods. A comparison on each method's energy consumption, cost and efficiency is also not a part of this work because of the complexity of advancements made in research on each method and process. Some methods have only been performed in laboratory scale and therefore in future research, more studies could be conducted in pilot to gauge the efficiency, energy consumption and cost.

AUTHOR CONTRIBUTIONS

The study was performed and written by C.P.M.; It was reviewed and edited by M.T., Y.P., and S.L.; Supervision by L.Z., Z.Z., and A.Z.

DECLARATION OF INTERESTS

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

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