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Recycling spent lithium-ion batteries: A holistic approach for addressing environmental challenges and resource recovery

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ABSTRACT

The globally projected share of Lithium-Ion Batteries (LIB) in the market will be around 875 million tons by 2025, leading to the generation of a tremendous amount of spent LIB trash to be dealt with. However, literature shows that only a tiny fraction of spent LIB is recycled currently, while the majority ends up in landfills, leading to environmental degradation. Though there is existing literature discussing the research trend and methods for recycling spent LIBs, very few reviews cover a comprehensive comparison of all the recycling methods along with the pretreatments. The major objective of the paper is to provide a comprehensive overview of the research landscape regarding LIB recycling, emphasizing the significant advancements in the field and valuable insights into the latest developments in LIB recycling technologies through a critical review of the recent and highly cited literature for spent LIB recycling. The paper focuses on three primary recycling approaches: pyro-metallurgical, hydrometallurgical, and direct recycling. The paper also covers major LIB types, analytical methods, spent LIB disposal challenges, the need for recovery of heavy metals, and pretreatment methods for LIB waste recycling. The paper further discusses the characterization techniques for leachates generated during hydrometallurgical processes, revealing the presence of various metals such as Al, Co, Cu, Fe, Li, Mn, and Ni. The detailed systematic review thus highlights the LIB recycling prospects and obstacles, and further research required to stimulate the creation of inventive and long-lasting solutions for a circular economy leading to sustainable development.

1. Introduction

Due to the growing need for replenishable energy resources and environmental concerns, power storage technologies, particularly batteries, have been the subject of extensive research [1]. Government regulations and market factors have accelerated the development and widespread use

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of plug-in hybrid electric vehicles (PHEVs) and battery electric vehicles (BEVs), intending to move away from fossil fuel-powered vehicles. Table 1 shows the exponentially increasing global stock of electric cars during 2013-2023 [2]. Electric vehicles mainly make use of lithium-ion batteries as their source of energy because of their high energy and power density, extended lifespan, and robustness. Changes in the LIB market share between 2017 and 2025 are depicted in Table 2 [3].

Global demand for LIBs is expected to rise 19 times over the next ten years, mostly due to the growing EV sector. After serving for roughly five to ten years, these LIBs will retire [4]. Mobile phones, which also use LIBs, possess a three-year lifespan on average and thus go in for recycling quicker than batteries used in EVs [5]. If we talk about India only, Figure 1 can give an idea about the share of lithium-ion battery waste by source [6]. LIBs present difficulties, and one dominant issue is the impact the production of batteries and their discarding operations have on the environment. End-of-life LIBs should be disposed of properly to avoid environmental contamination and fire hazards [7]. This presents difficulties because of the diverse chemistries involved and the safety risks of handling expended batteries [8]. At present, merely 5% of the used lithium-ion batteries (LIBs) are reprocessed using appropriate recycling and discarding techniques. The rest are either illegally reused, exported, incinerated, or dumped in landfills [9]. Mineral resources and brine are nowadays used in conventionally manufacturing lithium-ion batteries. A small group of nations (Australia, China, Chile, Canada, and Argentina) are home to almost 80% of the world's mining resources [10]. The global production of the major LIB materials of lithium, manganese, graphite, cobalt, and nickel are demonstrated in Figure 2a-e, respectively [11].

Table 1. Exponentially increasing global stock of electric cars during 2013-2023 as per 'Global EV Outlook 2024'.

Sr. No.	Year	No. of Electric Cars
1	2013	0.4 million
2	2014	0.7 million
3	2015	1.3 million
4	2016	2.1 million
5	2017	3.1 million
6	2018	4.9 million
7	2019	7.3 million
8	2020	10.1 million
9	2021	16.5 million
10	2022	26.3 million
11	2023	40.5 million

Table 2. Comparison between global share of LIB in 2017 and 2025.

Sr. No.	Device	Percentage of LIB used (2017)	Percentage of LIB used (2025)
1	EVs	57	75
2	Portable electronics	26	9
3	Industrial energy storage	5	5
4	Others	12	11

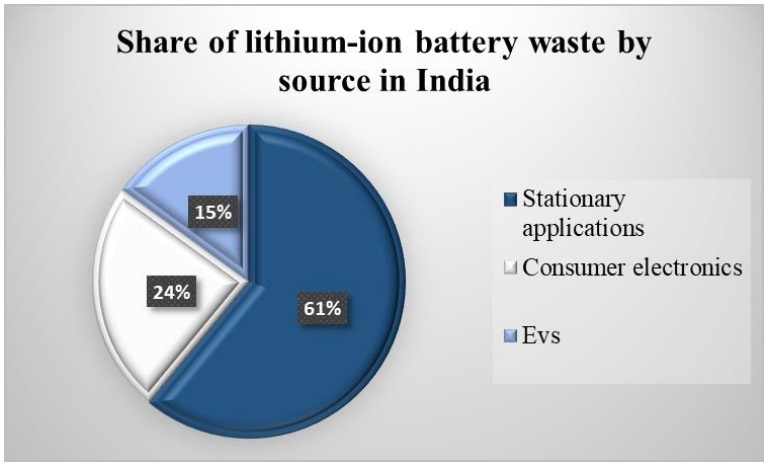

Fig. 1. Share of LIB waste in India.

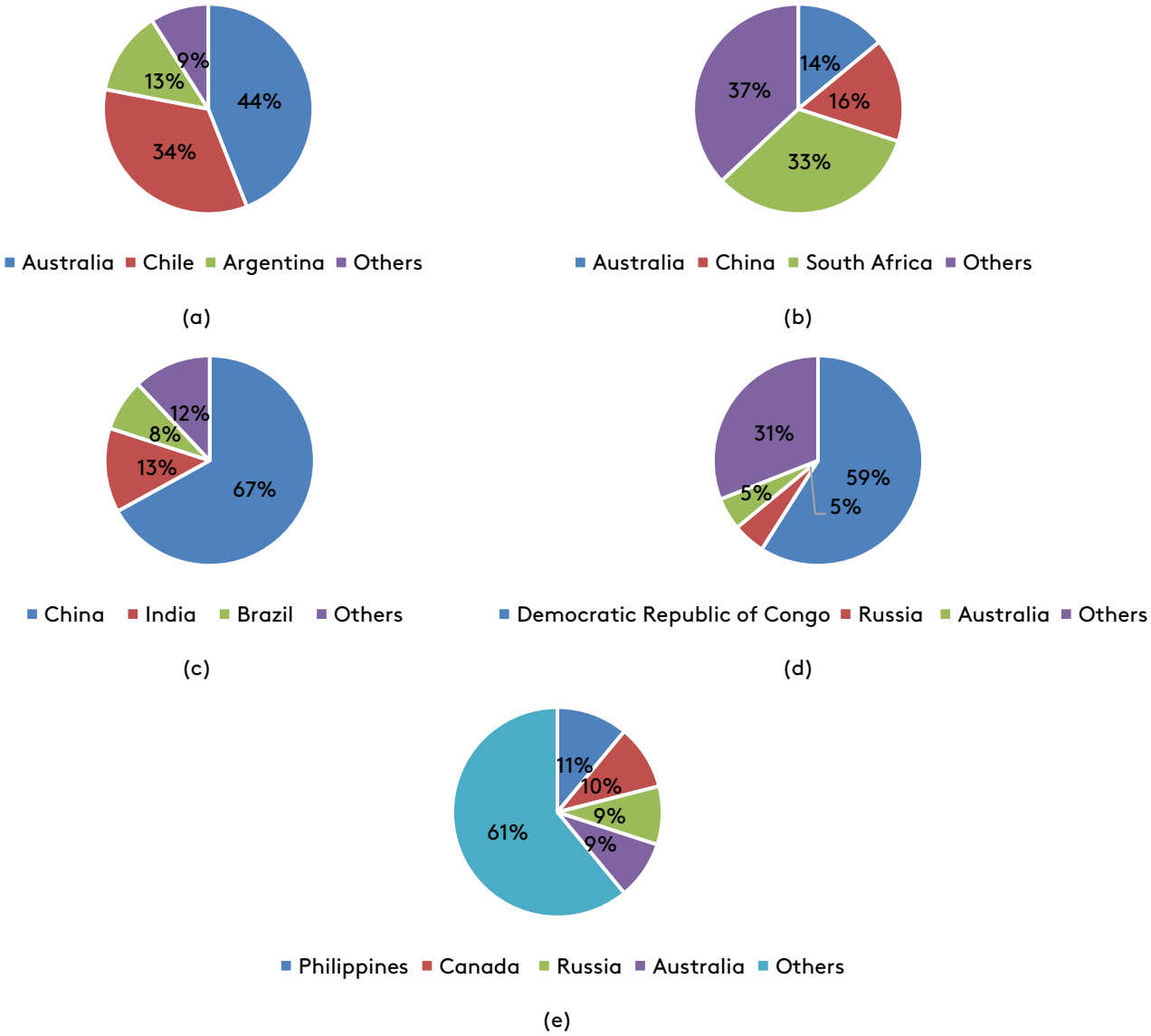

Fig. 2. Global production of (a) Lithium, (b) Manganese, (c) Graphite, (d) Cobalt and (e) Nickel

While lithium removal from saltwater or brines requires long vaporization periods, the extrication of lithium from its clays and minerals is energy-intensive and entails substantial mining expenses [12]. The compelling incentive-based politics of national legislation for growing the reclaiming of old battery trash may be explained by these strategic considerations combined with the growing demand [13]. After their useful lives are over, almost 95% of Li-ion batteries are disposed of in landfills rather than recycled. The restricted capacity of industrialized recycling methods only allows for the recovery of secondary raw materials, which are unsuitable to be used directly again in new batteries. Additionally, for most modern technologies to be economical, significant concentrations of the metal to be recycled are required, and thorough battery sorting is required before processing [14]. Most procedures waste the $FePO_4$ in lithium iron phosphate (LFP) batteries and only concentrate on recovering nickel, cobalt, manganese, and less lithium [15]. As a result, it's critical to develop suitable disposal strategies and recycling technologies to handle the consequences of discarded LIBs and the weakness in the supply chain of essential metals inside LIBs [7]. The huge quantities of precious elements in spent LIBs from EVs, like lithium nickel cobalt manganese oxide cathode, will make them an important secondary source [16]. Although lithium-ion batteries are a well-established technology, there are yet other ways to enhance their safety, cyclability, and performance. Significant changes in the extraction and applications of lithium are anticipated in the future [17]. There is enough room to eliminate the steps now involved in the process while simultaneously increasing the effectiveness of metal extrication and dissociation, encompassing lithium recuperation [12]. There is an urgent need for more quantitative frameworks, such as GABREAL, which is a supply chain model for analysis of the economic, environmental, and spatial aspects of recycling lithium-ion batteries in EVs, for the terminated automobile battery market in the UK. Additionally, there is a call for the creation of novel and more sophisticated techniques for material restoration from LIB waste. Such frameworks can provide details about the time when waste production levels will make a plant break even, as well as the location of the new plant or plants [18]. The United States, China, and Germany have the most supported projects in this field; nevertheless, developing nations are far behind in investment [19].

The recent review papers on the recycling of spent lithium-ion batteries provide critical insights into the current technological advances in the field. C. Dong et al. explained the reaction mechanism during various recycling methods and discussed the benefits and drawbacks of these methods to serve as a basis for pragmatic implementations. They also discussed the unexplored aspects of LIB recycling, such as material recovery from anodes and binders [20]. The recent progress in research and practical reprocessing of used ternary LIBs, including pretreatment of spent LIBs, pyrometallurgical recycling methods, hydrometallurgy, a hybrid of pyrometallurgy and hydrometallurgy, and direct recycling, was elaborated on by H. Liao et al. The paper also discussed the advantages and drawbacks of all these methods and addressed them based on their attainability and economic benefits [21].

The literature has also considered the working mechanism of each traditional recycling method, recent evolution in research and upcoming technologies, and new pretreatments for metal recovery, considering their odds, challenges, benefits, and disadvantages [22]. The costly resurrection of materials and the associated formation of tremendously poisonous gases linked with pyrometallurgical and hydrometallurgical systems of recycling ask for the development of substitute methods to circumvent landfilling waste batteries. K. Dhanabalan et al. discussed various eco-friendly, economically viable, and sustainable reprocessing methods for recycling divergent LIB cathodes that could be used to form a circular economy [23]. The literature also throws light on the foundation for understanding the theory of categorization of LIBs, pretreatment methods, and recovery of metals from electrolytes. The concept of direct regeneration and the mathematical calculations and simulations involved were elaborated on by P. Li et al. They concluded that the principles of physical chemistry are crucial for effectively separating the different elements of LIBs. [24]. An overview of various aspects of LIB

recycling, from the collection of raw material to metal recovery, targeted investigation of the mechanism of reclaiming all the components of LIBs and optimizing the processes for arriving at an improved system in the future. Life cycle assessments were also done to confront the problem of maintaining the supply chain of the battery industry [25].

The latest literature review papers discussed above and most of the previous literature review research papers have majorly discussed conventional recycling methods, like hydrometallurgy, pyro-metallurgy, and mechano-chemical processes, also referred to as direct recycling or regeneration methods. Most of the past literature reviews have provided discussions on the advantages and disadvantages of these methods.

The present paper provides a comprehensive overview and discussion of the literature on the characterization techniques for LIB leachate, a life cycle assessment of the entire recycling process, the impact of spent LIB on the environment, and a detailed discussion and overview of the integration of the recycling process with the circular economy, which is the novelty of the present paper. In addition, the latest research trend in the field of recycling LIBs through detailed scientometric analysis is discussed. The distinctiveness of the present paper also lies in the vastness of facets it collates about lithium-ion batteries. These chunks of information, though available in the existing literature, are seen only in bits and pieces.

The paper is divided into seven primary sections, the first being the introduction. The general classification of LIBs is discussed in the second section, along with their global percentage usage, their functions, the chemical composition of each unit, and the working of different components of batteries. This section explains why the current recycling methods focus mainly on recovering metals from the cathodes of LIBs. Section three emphasizes treating spent LIBs as a substantial source of metals essential in replicating such batteries. The impact of improper disposal of spent LIBs is discussed in section four. In section five, the need for recovering metals from used LIBs is elucidated. Section six introduces the pretreatment methods for metal retrieval. Section seven concentrates on metal recovery and the benefits of recovering precious metals from LIB leachate. This section also includes various characterization techniques used for studying LIB leachates and gives the general composition of LIB synthesized in the lab during some earlier research works. The paper aims to establish a comparison between the methods currently used for recycling LIBs and to serve as an important resource for researchers inclined towards LIB recycling.

The critical metals crucial for battery production are concentrated in limited regions of the world, and the resources are also limited. Based on resource availability and the requirement for specific purposes, various battery chemistries are developed. The section below discusses lithium-ion batteries' types, composition, and basic workings.

2. Classification, components, and working of LIB

2.1. Classification of lithium-ion batteries

LIBs are often classified into six types based on the material used to produce the cathode, which includes lithium nickel cobalt aluminum oxide (NCA), lithium cobalt oxide (LCO), lithium iron phosphate (LFP), and lithium nickel manganese cobalt oxide (NCM) [26, 27]. Their characteristics and applications are dependent on the cathode material used [27]. Table 3 provides information on the types, composition, applications, advantages, and disadvantages of different battery types. Figure 3 shows a comparison of the percentage of different LIBs in 2017 and that estimated to be used globally in 2025 [3]. From the figure, it can be observed that the usage of NMC increased over time, and that of the FLP batteries dropped because of its heaviness, which presents installation difficulty. Also, LFPs have low energy density and low voltage, which are again overcome by NMC batteries [16, 27-30].

Table 3. Types of LIBs, their composition, applications, advantages and disadvantages.

Battery Type	Cathode Components (Li being common in all)	Life cycle	Application	Advantages	Disadvantages	Reference
LFP ($LiFePO_4$)	Lithium (4%), Iron (35%), Phosphate (61%)	2,000 - 5,000	Short-range electric vehicles, energy storage systems	Iron is cheaper, easily available, has high-temperature stability and cyclability, and is more environmentally friendly than cobalt and nickel. Have a long-life span. Safe.	Heavier and pose a problem during initial installation. Low energy density. Low voltage.	[16, 27-29]
NMC ($LiNiCoMnO_2$)	Lithium (11%), Nickel (30-80%), Manganese (8-28%), Cobalt (9-31%)	1,000 - 2,000	E-scooters, Electric buses, transportable PCs, power equipment, systems for storing energy	High energy density and power density, improved safety	Co and Ni both are costly and detrimental to the environment, with poorer cycling performance compared to LCO	[16, 27, 28, 30]
NCA ($LiNiCoAlO_2$)	Lithium (11%), Nickel (75%), Cobalt (14%), Aluminium	1,000 – 2,000	Portable electronics, laptops, smartphones, power banks, long-range EVs, tablets	Greater capability, extended shelf life, less expensive than LCO	Prone to thermal runaways. Poor cycling performance, production difficulty	[16, 28, 30]
LMO ($LiMn_2O_4$)	Lithium, Manganese	500 - 800	EVs, power tools, portable electronics, medical equipment, energy storage systems	Slightly cheaper manufacturing cost. Function well at higher temperatures. Charge quickly. Have high voltage.	Poor cycling performance, low capacity	[16, 27]
LCO ($LiCO_2$)	Lithium (11%), Cobalt (89%)	500-1,000	Laptops, smartphones, tablets, digital cameras, portable electronics	High energy density, less self-discharge, excellent cycling efficiency, simple manufacturing	Poor heat stability. Short lifespan, high cost	[16, 27, 28, 30]
LTO (Li_2TiO_3 / $Li_4Ti_5O_{12}$)	Lithium, Titanium	Up to 10,000	Electric powertrains, UPS, solar-powered street lighting, supercapacitors	Long life cycle. Less polluting than other alternatives. Charge quickly.	Low energy density. Expensive.	[28]

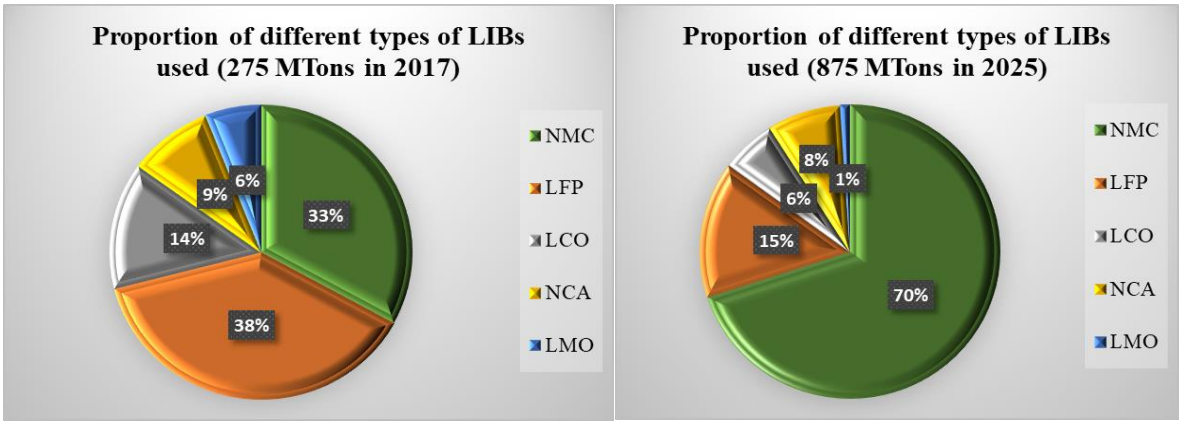

Fig. 3. Comparison of the percentage of different LIBs in 2017 and that estimated to be used globally in 2025.

2.2 Working mechanism of lithium-ion batteries (LIBs)

The major components of a lithium-ion battery are an anode, a cathode, a separator, an electrolyte, and two current collectors [31]. Figure 4 shows the average composition of the components by weight percentage in LIBs [32]. Similar observations are found in other sources as well, where graphite (anode) forms 16%, cobalt, lithium, manganese, and nickel (cathodes) together form 23%, aluminium collectors form 15%, and copper forms 10% of lithium-ion batteries [33]. The separator, anode, and cathode are submerged in liquid electrolyte and sealed within a plastic bag/aluminum box/stainless steel container. Graphite powder is the active ingredient in the anode. It is combined with a binder, usually polyvinylidene fluoride (PVDF), and coated on a Cu foil current collector. Lithium metal oxides or phosphides, such as $LiCoO_2$ (LCO), $LiMn_2O_4$ (LMO), $LiNi_xCo_yMn_zO_2$ (LNCM), $LiNi_xCo_yAl_zO_2$ (LNCA), and $LiFePO_4$ (LFP), can be included in the cathode. These materials are layered on a carbon black-equipped aluminum current collector, which acts as the conductor, and the binder in the form of PVDF [34]. Both the anode and the cathode contain lithium. The lithium ions having a positive charge are borne by the electrolyte from the positively charged electrode (i.e., anode) to the negatively charged electrode (i.e., cathode) and vice versa via the separator. Li ions shift from the anode side towards the cathode when the battery discharges, producing the flow of current because electrons are produced. Once the batteries have been charged via a separate power supply, the opposite happens [35]. The electrolyte facilitates the passage of Li ions within the cell, while a voltmeter measures the electric current flowing via the external connection [27]. Figure 5 demonstrates the working of a Li-ion battery [32].

While charging, the flow of lithium occurs towards the anode, thus changing the voltage potential. These lithium atoms start forming a solid electrolyte interphase (SEI) layer on the anode surface and hinder the conduction of ions, thus diminishing the battery capacity. This SEI layer, formed of lithium oxide and lithium carbonate, expands in size with the increasing number of cycles. The cathode also encounters a similar film called the electrolyte oxidation layer. Ultimately, batteries die as they cannot contain charge after a particular number of cycles. Such batteries that become completely degenerated and their further charging is not possible are called spent LIBs. Although these batteries cannot serve their original purpose of providing power, they can still act as a prominent source of critical metals [36].

3. Spent lithium-ion batteries (S-LIBs): a significant mineral resource

End-of-life (EOL) LIBs have surged due to the quick rise in their use in electric vehicles and portable electronics [37]. The rapid expansion of Li-ion battery commercialization has put a heavy strain on metal mineral suppliers, especially those that supply the rare element 'lithium' [38]. In addition, numerous important metals (Li, Mn, Cu, Co, Ni, and Al) can be found in the used Li-ion batteries. Consequently, used LIBs are considered "artificial minerals" in the extraction of metals [39]. Since the battery cell compositions differ significantly

from one another, a cell chemistry-specific strategy is required to maximize the environmental benefit and output quality during recycling [40]. As a result, developing an effective recycling method for spent LIBs has received huge attention for reasons related to the environment and the economy [37]. From an economic perspective, waste lithium-ion batteries (LIBs) are a valuable resource containing elements including cobalt, manganese, and nickel [41]. Figure 6 shows the materials each component of LIBs is made up of [42-46]. Numerous studies have been conducted on the reuse of precious metals from wasted LIBs. The recovery techniques frequently employed include extraction, crystalline hydrometallurgy, pyrolysis, pyrometallurgy, and mechanical grinding [47]. Such techniques are, however, constrained by difficult procedures, secondary pollutants, and expensive production costs [48]. Finding more effective, reliable, and long-lasting techniques for recycling and treating wasted lithium-ion batteries is therefore essential. A detailed literature review of methods and their suitability is provided in section 7.

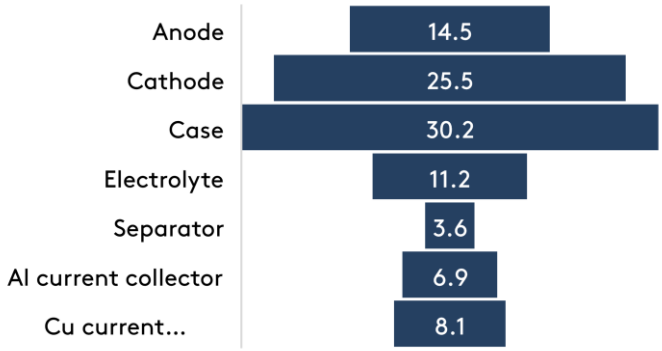

Fig. 4. Average percentage of different components of lithium-ion batteries.

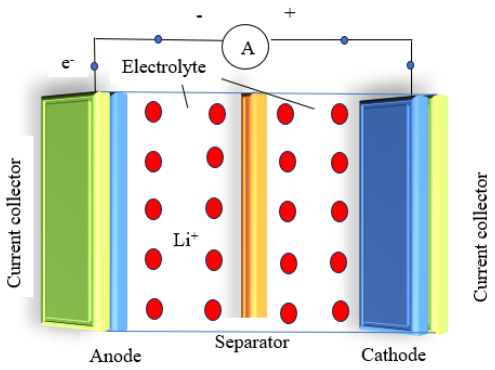

Fig. 5. Working of LIB.

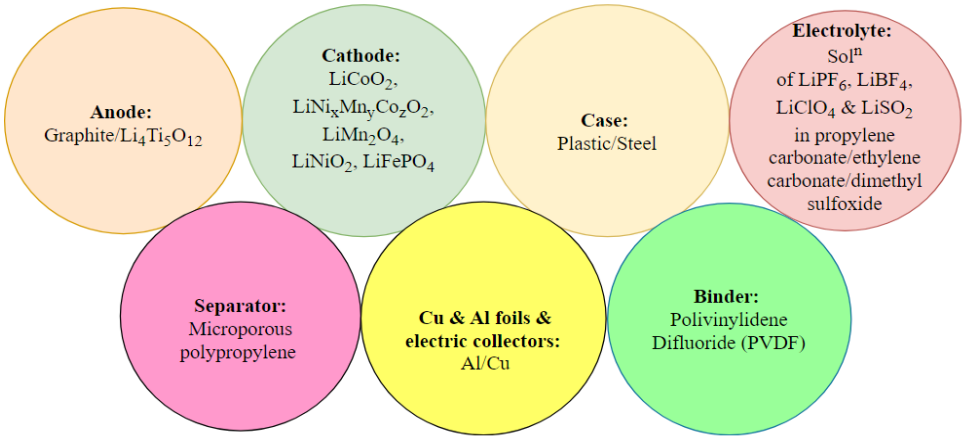

Fig. 6. Components of LIBs and materials commonly used to make them.

4. Necessity for metal recovery

The typical lithium-ion battery composition includes lithium (5-7%), cobalt (5-20%), and nickel (5-10%). Additionally, 5–10% of the battery incorporates a range of complementary heavy metals, like copper, aluminium, iron, chromium, etc. [10]. Because of their composition, waste materials from Li-ion batteries present an amazing alternative as a mining resource [13]. Additionally, other predominant sources of battery waste are mobile phones, which contain a variety of metals, including antimony, beryllium, palladium, zinc, copper, lithium, manganese, cobalt, gold, silver, nickel, and rare earth metals [49, 50]. Many academic studies have been centered on the recuperation of active portions of batteries since reusing old lithium-ion batteries for recycling has grown in importance in contemporary culture [51]. The majority of the benefits to the environment come from the recuperation of battery pack Al and Cu fractions, with prominent additional benefits

coming from the retrieval of battery cells Ni and Co [52]. Moreover, it has also been demonstrated that reusing trash LIBs uses a smaller amount of energy and resources than creating fresh ones made of virgin materials [27].

The potential environmental effects of producing LIB cells can be significantly reduced through recycling; as such, recycling is essential for the future of the LIB industry. This was discovered with the help of a cell-specific assessment of LIB and post-LIB cell recycling using various recycling processes [40]. Based on the ReCiPe characterization approach, another Life Cycle Assessment (LCA) model demonstrated a net advantage in 11 of the 13 environmental effect categories when compared to battery production without recycling [52]. It is still debatable if remanufacturing LIBs using recycled elements from old batteries has environmental or financial benefits; thus, more research is necessary. A study was carried out to evaluate the emissions of greenhouse gases (GHGs), usage of water, and related expenses for LIB remanufacturing using a life-cycle framework and a process-focused costing model with four different LIB kinds (NCM_{111}, NCM_{622}, NCM_{811}, and NCA). The findings indicate that, in comparison to producing LIBs using virgin materials, remanufacturing LIBs can lower production costs, water usage, and greenhouse gas emissions [53].

From the perspective of environmental sustainability, the recycling of spent LIB and the recovery plays a major role. The critical metals, such as lithium, copper, cobalt, manganese, and nickel, used in manufacturing LIBs remain intact in their spent masses. When LIBs are not sent for recycling but are inappropriately disposed of in landfills, these heavy metals have been found to contaminate each aspect of the environment, including soil, water, and air. Preeti Mishra and Sayali Apte (2025) investigated experimentally how heavy metal concentrations in clayey soil affected their soil index and engineering properties of the landfill liner by neutralizing electrostatic forces between the particles of soil due to metal ion adsorption [54]. While standing stagnant as landfill deposits, the waste batteries may come in contact with various liquid environments, either through rains or surface waters, thus forming leachate. Leachate corrodes the battery materials over time, therefore bringing the heavy metals in contact with ground and surface water and polluting them [55]. Also, fragments of heavy metals escaping from inappropriately closed landfills can affect the respiratory tracts of humans and animals. Even recycling operations generate air contaminants in the form of dust and acidic or organic gases [56].

5. Impact of improper disposal of spent LIBs

From the literature discussed above, it is evident that only a small fraction of the total trash LIB generated undergoes recycling. The remaining portion is usually dumped in landfills. Health risk assessments done in the past have proved the presence of heavy metals in drinking waters at hazardous levels. For instance, a study showed the target hazard quotient (THQ) and hazardous index (HI) figures regarding arsenic and the target cancer risks (TCR) for humans in maximum test samples surpassed the maximum risk limit (i.e., THQ < 1.0; HI < 1.0; and TCR = 10^{-4}). Thus, necessitating the frequent monitoring of drinking waters to ensure the safety of the consumers [57]. Spent LIBs can be significant sources of some of these heavy metals. Inappropriate or reckless spent battery processing and disposal contaminates the land, water, and air. If LIBs are disposed of on land, rains will carry the contents into the surrounding soils, where they may seep into deeper layers, contaminate groundwater, and possibly runoff into surface waters [58]. The literature indicates that this LIB leachate contains metals like aluminium, cobalt, copper, iron, manganese, and nickel. The fact that LIBs contain a significant amount of hazardous heavy metals makes them a highly distinct concern [59].

Such heavy metal contamination in water is dangerous to aquatic life as well as human health. Furthermore, these heavy metals find their way into the food chain by way of the soil, affecting the well-being of every living being. There is also a possibility that surface fires in landfills will move outside the boundaries of the landfill. Methane gas, which is produced in and moves through an MSW dump, has the ability to intensify such fires [60]. When expended lithium-ion batteries are not disposed of properly, it can lead to serious issues

like precious resource wastage and environmental damage [61]. Table 4 briefs the effects spent LIBs can have on various aspects of the environment.

Recycling LIBs addresses two major issues: 1) adverse effects of LIB production on the environment and 2) proper management of spent LIB garbage [53]. Thus, recycling of LIBs must be promoted to achieve environmental sustainability and resource conservation, and the use of recycled metals should be endorsed in the manufacturing of the battery. The recycling of spent LIB involves an elaborate pretreatment followed by the actual recycling. The following section provides an in-depth discussion of the methods of pretreatment and a critical analysis of the various methods used and their suitability.

Table 4. Impact of LIB on the environment.

	Environmental aspect	Impacts	References
Impact on	Soil	Dissolved gases (HF, HCl, SO_2) in LIB leachate cause acidification and make soil corrosive.	[58]
		Plants absorb lithium, thereby introducing Li to the food chain.	[62]
		High Li concentrations inhibit plant growth.	[62, 63]
		Impacts plant metabolism.	[62, 64]
		Lithium toxicity lowers the amount of chlorophyll.	[62]
	Water	Elevated Li concentrations in water are detrimental to aquatic life.	[62]
		Landfill fires carry particulate matter and chemicals to nearby water bodies and raise the Chemical Oxygen Demand (COD).	[60]
		Cell apoptosis in human cardiomyocytes is stimulated.	[65]
		Li harms benthic organisms, on which the aquatic ecosystem's survival depends.	[62]
	Air	When a landfill fire occurs, smoke and hazardous gasses are released.	[58, 66]
		LIBs discharge fine particles (metals from the breakdown of LIBs, electronic debris, including As, Cd, Cr, Co, Cu & Pb) into the atmosphere during the disassembly and recycling operations.	[67-69]
		Dust causes heart & lung disorders, cancer, endocrine disruption, and other health effects.	[58, 62]
		HF entering the human body through the respiratory or cutaneous systems has a corrosive and poisonous effect on the entire body.	[58]
	Other impacts	LIBs release electromagnetic radiation both during and beyond their useful lives.	[50]
		Li at high levels (15–20 mg/L blood concentrations) causes nausea, visual impairment, kidney issues, cardiac arrest, and coma.	[62]

6. Waste preparation or pretreatment

In order to reduce the dangers involved in handling and manipulating LIBs because of their residual stored energy, the first treatment step focuses on battery deactivation [70]. Generally, the discharge involves immersing LIBs in solutions that are saturated with either NaCl or Na_2SO_4, minimizing the risk of explosions by reducing the amount of metallic lithium [70, 71]. Using aqueous salt baths may also cause internal components to get contaminated with undesirable ions and produce

harmful hydrogen fluoride (HF) and other gases due to the electrolyte leaking into the water, especially in the case of high-voltage battery packs. Alternatively, LIBs might be connected to resistors to gather and utilize any leftover power, or they could be covered in chips made of stainless steel to initiate a planned short circuit [72]. Dismantling also attempts to reduce the volume of the product that needs to be handled, especially in the case of EV LIBs [70].

Pretreatments can be of three types: thermal, mechanical or physical, and chemical or mechanochemical.

6.1. Thermal pretreatment

The organic components of LIBs are most affected by high-temperature treatments. The primary obstacle to material separation may be the binder, which is made up of Polyvinylidene Fluoride (PVDF) and is accountable for the adherence of the active powder to the foil collectors used currently [70]. A SEM study verified the ideal temperature range for its breakdown, which was determined to be 500–600°C [73]. Although elevated temperature thermal procedures can increase the effectiveness of lithium recovery by 90%, they also produce considerable emission of harmful gases (e.g., dioxins, hydrogen fluoride, carbon monoxide, carbon dioxide, etc.) that necessitate the application of gas scrubbers and air-filtering systems [74].

6.2. Mechanical or physical pretreatments

The most widely used industrial strategy combines pretreatments, both physical and mechanical, to remove the outer casing, separate important elements, and minimize scrap volumes. The potent powder separates from the base, the clusters break apart, and two or even more fractions are produced because of the impact strains that are produced while grinding, which transform kinetic energy into energy responsible for the breakage [75]. The greater surface area will have an impact on the following hydrometallurgical process, encouraging the dissolving of metals during acid leaching. Usually, an external steel casing is removed by magnetic separation in the initial crushing stage. Fine grinding comes next to remove current collector foils and powdered organic ingredients that are actively leachable [70].

6.3. Chemical or mechano-chemical pretreatments

Chemical pretreatments involve the extraction of the electrolyte or the dissolution of the binder using hydrocarbon-based solvents and supercritical fluids like CO_2 [70]. It has been demonstrated through testing that powerful bases and acids, hydrocarbons, halogens, and powerful oxidants don't react with the PVDF binder; however, solvents that are organic, like N, N-dimethylformamide (DMF) or N-methylpyrrolidone (NMP), exhibit high solubility with the binder [76]. However, more recent research also looks at less expensive and more environmentally friendly substitutes, such as juice from citrus fruits or the Hill Lemon Galgal treatment, which can disintegrate the PVDF binder [77]. The primary drawback of these pretreatment processes is their strong reliance on chemical makeup. Lastly, mechano-chemical pretreatments benefit from the synergistic action of leaching metal chelate agents like PVC and ethylenediaminetetraacetic (EDTA) and grinding. The polymorphic change and bond breaking of the cathodic material are brought on by the reduction in particle dimensions and an increase in the area of the surface, which boosts the reaction process for the following acid-leaching process [78].

As seen in the literature, the precautionary step of pretreatment is essential to dispense with the enduring stored energy in used LIBs, their tendency to explode under pressure, or chemical reactions involved in further recycling processes, and to achieve volume reduction. The three existing pretreatment methods, though capable of eliminating different components of LIBs, are associated with various drawbacks. For instance, thermal pretreatment effectively removes organic components such as the binder but also yields detrimental gases, thus increasing the cost by compelling the use of gas-tackling mechanisms. Combining mechanical and physical forces to bring about a size reduction of LIB trash, despite leading to good recycling performance, causes the escape of fine metal dust into the environment in the absence of a closed, controlled environment, which is harmful to human respiratory health.

Chemical solvents, which do not produce gaseous contaminants, can dissolve electrolytes and the binder, sometimes even without reacting with them. However, using chemicals may prove

expensive and prompt the production of hazardous secondary wastes. The available literature suggests modification of the existing pretreatment methods or the development of new ones that are cost-effective and environmentally harmless. Thermal pretreatment should be restricted to such temperatures up to which good quality recycled metals can be obtained without releasing toxic gases and energy consumption is minimized. Mechano-physical methods followed by the segregation of individual battery components should be preferred to avoid the pain of obtaining pure metals from the recycled products later. Solvents that are benign to the environment and that produce no harmful products with the battery materials should be selected for the chemical pretreatment method. The pretreatment of the spent LIB is followed by recycling and recovery of the critical elements. The complex battery construction and electrode material composition necessitate physical and chemical/electrochemical procedures for LIB recycling and metal recovery. The section below critically analyses the various methods for recycling and the areas of further research leading to sustainability and circular economy.

7. Methods of recycling and metal recovery

Physical processes include pretreatments like washing, magnetic separation, disassembly, crushing, screening, and heating. Chemical processes encompass a variety of techniques, like the extraction of solvents and chemical precipitation, bioleaching, acid leaching, and electrochemical processes [37, 59, 61]. The variety of raw materials makes it difficult for a single technology to be economical as well as ecologically benign. For this reason, recycling wasted LIBs is commonly done by combining physical and chemical methods [72]. Figure 7 shows a schematic illustration of the steps followed for material recovery from spent LIBs [37].

Currently, pyrotechnical, hydrometallurgical, and direct recycling are the three primary methods used to treat spent LIBs [79].

7.1. Pyrotechnical or pyrometallurgical recycling

Rechargeable battery recycling usually begins with a mechanical procedure that can greatly improve recovery and extraction using pyrometallurgical techniques. Mechanical pretreatments include the generally manual process of sorting, as well as automatically performed crushing, grinding, and screening [80]. Through high-temperature calcination, the sustainable retrieval of laminar $LiCoO_2$ components from used cell phone batteries was investigated. From the study, it was determined that active substances could be effectively recuperated from used mobile phone batteries by separating them from the aluminium thin foil and then calcining them at a high temperature [81].

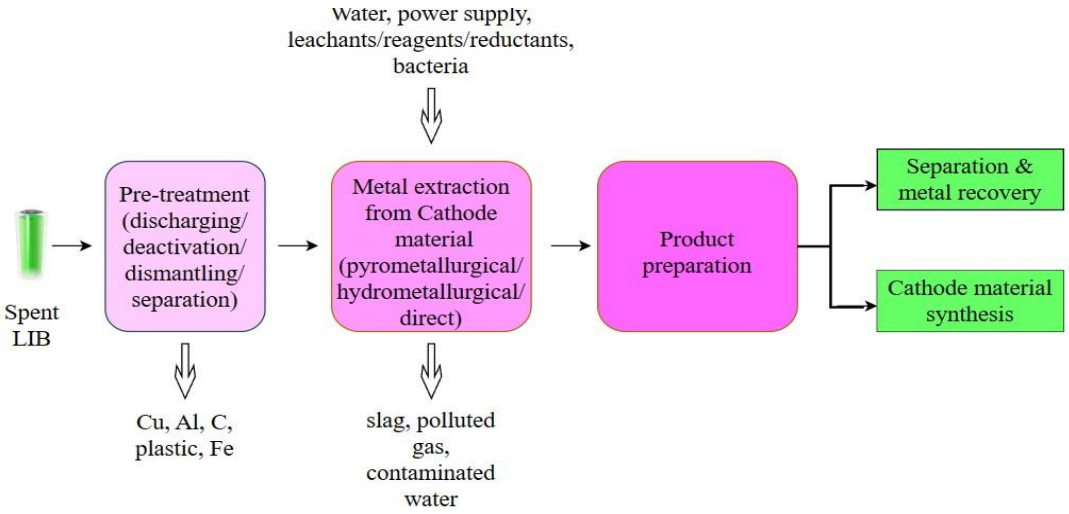

Fig. 7. Steps for spent LIB recycling.

Cost-neutral and sustainable recycling using pyrometallurgical recycling in conjunction with mechanical pretreatment was done to recover all battery components, particularly Li from powdered electrode mass, which is a tiny portion physically removed from used Li-ion batteries. Two methods of metallurgical treatment were examined: lithium oxide recovery with selective entraining gas evaporation, and direct vacuum evaporation of lithium and distillation to extract the metal lithium [82].

The organic electrolyte, binder, and acetylene black found in the discarded LIBs are frequently burned out using pyrometallurgical procedures, resulting in significant energy usage and the production of toxic gasses [83].

It is possible to establish a coupling process that decreases the temperature at which LCO decomposes from 1426 K to 1173 K by burning the positive material graphite and addressing the negative material to the pyrolysis process. Carbon's (in graphite) strong attraction to oxygen can destroy oxygen octahedrons. Li and Co can readily escape as O octahedrons collapse, which supports the reaction to continue at a relatively low temperature [84].

The global warming potential (GWP) and cumulative energy demand (CED) of three pyrometallurgical technologies were analyzed and compared: an emerging direct-current (DC) plasma smelting technology (Sc-1), the same DC plasma technology but accompanied by an extra pretreatment stage (Sc-2), and a more developed commercial ultrahigh temperature (UHT) incinerator (Sc-3). The findings showed that, when pretreatment was used, as is the case with Sc-2, transferring to DC plasma technology from UHT furnace technology (Sc-3) might reduce the recycling process's greenhouse gas emissions by up to a factor of five. According to open-loop recycling assumptions, just in Sc-2, the emissions of carbon and energy-related credits provided by the metals that were retrieved compensate excessively for energy and emissions needed to complete the battery recycling procedure, leading to overall values that are negative for both GWP and CED [85].

A basic comprehension of the thermokinetic reactions involved in the recycling of LIBs through pyrometallurgy is necessary to develop an optimum sustainable recycling process. Therefore, the kinetics and thermodynamics of processes at high temperatures for LIBs while recycling using pyrometallurgy were examined by means of a thermogravimetric analyzer (TGA), having a gas mass spectrometer attached. A consistent heating rate of 20°C/min was applied to composite pellets that were cylinder-shaped, containing active materials for the cathode and anode until they reached 1500°C. The predicted theoretical thermodynamic processes were ascertained by contrasting the mass loss and concomitant gas evolution measurements. Temperature-sensitive processes and quicker mass transfer were revealed by isothermal decreases from 850°C to 1000°C [86].

During the smelting process, active metals like lithium and aluminum that have strong reducibility are susceptible to oxidation and end up as their oxides in the smelting slag [87]. Thus, after heat treatment, lithium, aluminum, and all plastic components are lost in various pyrometallurgical processes [40]. Hence, pyrometallurgical techniques are unfit to extract Li as a useful element and are ineffective for batteries made of phosphate [15]. Since the InduRed reactor concept does not have this drawback, it may be a potential new method. The behavior of black mass from a pretreatment process under reducing conditions and nickel-rich cathode materials ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ and $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$) has been studied using differential scanning calorimetry, thermogravimetric analysis, and heating microscope experiments to demonstrate its suitability for black mass processing. Reaction temperatures of 800°C to 1000°C were theoretically achievable, and Mn, Co, and Ni had high recovery potentials. A Li elimination rate of up to 90% relative to its starting mass was accomplished, and the slagging of lithium was mainly avoided [88].

It is also important to look at the recent developments that have been made in this aspect. The section below discusses the recent developments in the pyrotechnical method for dealing with spent LIBs or LIB leachate.

A thermochemical approach has been suggested in the literature for managing plastic debris generated from spent batteries while using conventional recycling techniques (incineration,

landfilling, & pyrometallurgical/hydrometallurgical processes). The LIB separator served as the model feedstock. Co-feeding CO_2 at high temperatures (>500°C) enhanced the generation of syngas and C_{1-2} hydrocarbons with a decrease in liquid pyrolysates [89]. Resonant acoustic vibration (RAV)-based methods have shown up to a 92% stripping efficiency. Combined with heat treatment, RAV offers unmatched scalability and efficiency over traditional stripping techniques like curling-uncurling, sonication & magnetic stirring, doing away with the requirement for post-processing. The resultant cathode powders are pure and have their original polycrystalline structures [90]. For pyrolyzed black mass, two recycling processes, namely an early-stage lithium recovery using a hydrometallurgical technique and a pyrometallurgical method, were techno-economically assessed. This method aims to pre-separate Cu and Al using pyrolysis to maximize recycling efficiency. The hydrometallurgical recycling technique was projected to be more economical, as it turned out to be less vulnerable to cost variations and alterations in the chemistry of cells [91].

An alternative to the high energy use of the pyrometallurgical approach is an elevated temperature procedure powered by a more environmentally friendly energy supply, for instance, concentrated solar power. The battery trash comprising different cathode metals was recovered using anode carbon and carbothermic reduction in a solar simulator furnace. A thermodynamic evaluation was performed using a thermochemical package and a selection of experimental data obtained between 400 and 800°C [92]. By lowering the direct CO_2 emissions in pyrometallurgical processing, recycling LIB cathode materials with hydrogen, which is a cleaner reducing agent, would lead to a safer method of producing metal. With a thorough microstructure evolution observation and an isothermal mass change analysis, this work examined the rate at which $LiCoO_2$ reduced H_2 at 600–1000°C. The shrinking-core model seems to be the process of general reduction [93].

Despite the numerous benefits offered by this method, environmental issues could arise from the pyrometallurgical recycling process regarding photochemical ozone formation, global warming, and consequences that are both carcinogenic and non-carcinogenic, depletion of the ozone layer, and eutrophication [58]. The burning of the electrolyte and binder consumes a huge amount of energy and produces poisonous gases. Lithium and aluminium show high reducibility under heat and lose their existence to their oxide forms, and hence cannot be recovered using pyrometallurgy, especially in the case of phosphate batteries.

Some efforts can be taken to reduce the downsides of the pyrometallurgical system of recycling LIBs. The global warming potential, cumulative energy demand, and thermokinetic reactions associated with pyrometallurgical recycling systems should be closely monitored to observe an inoffensive and optimally sustainable recycling process. Combining the pyrotechnical method with physical processes has been found to eliminate the need for post-processing. Pre-processing for selective removal of Cu and Al improves the recycling capacity. The high energy required for pyrometallurgical treatment can also be obtained from environmentally benign solar power, thus making the system more sustainable.

7.2 Hydrometallurgical recycling

7.2.1 Techniques for characterization of LIB leachate

For recovery purposes, leachates are generated using various solvents alone or in combination: hydrochloric acid (HCl), hydrogen sulphide (H_2SO_4), D2EHPA –Bis (2- ethylhexyl) phosphoric acid, Bis 2,4,4-trimethylpentyl phosphinic acid, kerosene, $Na_2C_2O_4$, sodium hydroxide (NaOH), sodium carbonate (Na_2CO_3), DTAB –Dodecyl trimethyl ammonium bromide, hexane, nitric acid (HNO_3), organics acids (like citric acid, tartaric acid, malic acid, oxalic acid), ethanol (C_2H_5OH), etc. [47, 76, 94, 95]. To dissolve PVDF, which is the most commonly used binder material, N-methylpyrrolidone (NMP) is usually employed, since it serves as a good solvent for PVDF [76, 95 ,96]. Some environmentally benign solvents known as Deep Eutectic Solvents (DES) are emerging as effective replacements for conventionally used organic and inorganic solvents [97].

7.2.2 Composition of LIB leachate

Leachate formation occurs when the components of lithium-ion batteries come in contact with some compatible solvents. For experimental purposes, many solvents have been used in the past for leaching metals into them. Deionized water has also served as a solvent in some cases [98]. The leaching agent alone is sometimes not enough and some reducing agents like H2O2 have to be added along with these leaching solvents [41]. The general compositions of simulated leachates during different studies have been discussed [99, 100] and are presented in Table 5. Characterization techniques have been used alone as well as in combination with each other: inductively coupled plasma (ICP), microwave-induced plasma, atomic absorption spectrophotometer (AAS), X-ray diffractometer (XRD), X-ray fluorescence spectroscopy (XRF), scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FT-IR), X-ray photoelectron spectroscopy (XPS), etc. Table 6 shows the technologies used so far for the detection of different metals.

Hydrometallurgical methods are typically used for metal recovery. These methods involve a leaching step to dissolve the target element, followed by a separation step that can be accomplished by employing a variety of methods, including ion exchange, solvent extraction (SX), and precipitation. Hydrometallurgical metal recovery is often carried out at low temperatures [112]. Compared to the pyrometallurgical process, it offers numerous benefits, including low energy consumption, minimal emissions of dangerous gases, high extraction efficiency, and cheap capital costs. It has enormous industrial realization potential [72]. Because hydrometallurgical processing recovers more lithium as hydroxide, it is demonstrated to be more ecologically friendly than pyrometallurgical processing, which is expected to possess a bigger Global Warming Potential (GWP) because of its increased energy density [52]. Sulfuric, hydrochloric, and nitric acids are among the inorganic acids that are dissolved in the discharged, dismantled, crushed, and sorted expended lithium-ion battery black mass in typical hydrometallurgical processes, typically in conjunction with a reducing agent like hydrogen peroxide. Subsequently, the leached metals undergo selective recovery and purification by procedures like precipitation, crystallization, or extraction of solvents [47, 109]. The selection of suitable hydrometallurgical procedures is influenced by the elements present in both the waste and the targeted metals. Utilizing end-of-life Waste Electrical and Electronic Equipment (WEEE) as an alternative source of essential raw materials will become easier with a solid grasp of the bulk chemical composition of the WEEE (which includes LIB), leaching techniques, and the metal recovery process [113].

Table 5. Composition of various materials present in simulated LIB leachate.

Metal	Quantity (g/L)
Al	1.16-1.5
Co	11.91-17.8
Cu	1.48-2.2
Fe	0.34-0.94
Li	2.4-4.35
Mn	1.82-2.24
Ni	1.44-2.0

7.2.3 Leaching, precipitation, and extraction techniques

The most important stage in the hydrometallurgical process is leaching the different precious metallic species from the negative electrode into a solution because it is only in this form that the subsequent separation and recycling can be conveniently accomplished [34]. It is a common practice to use inorganic acids as leaching agents. For instance, Co, Mn, and Ni were extracted from LIBs using the leaching technique. Five grams of powder were used in the experiments, which examined the consequences of the concentration of sulfuric acid, H_2O_2 content, pulp density, and duration of leaching on the leaching processes. Inductively Coupled Plasma (ICP) analysis findings revealed that leaching of Co, Mn, and Ni at a pulp density of 50 g/L with 1 M sulfuric acid and 15% H_2O_2 (v/v) had great efficiency and simultaneously leached more than 99 wt. % of metals [41].

Table 6. Metals in leachate and their detection techniques.

Sr. no.	Methods	Metals detected	References
1	Inductively Coupled Plasma Mass Spectrometry (ICP-MS)	Co, Mn, Ni, Li, Al, Fe, Cu	[41, 98, 101, 102]
2	Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES)	Li, Ni, Co, Mn, Al, Fe	[38, 97, 103-105]
3	Fourier transform infrared spectroscopy (FT-IR)	Li, Co, Ni, Mn, Cu, Gr	[38, 39, 47]
4	X-ray photoelectron spectroscopy (XPS)	Li, Co	[39, 106]
5	X-ray diffraction (XRD)	Co, Li, Fe, Ni, Mn	[31, 38, 81, 98, 104, 106, 107]
6	X-ray fluorescence spectroscopy (XRF)	Cu, Li, Mn, Al, Co, Ni	[108]
7	Scanning Electron Microscopy (SEM)	Li, Co, Ni, Fe	[38, 39, 81, 104, 106]
8	Thermal Gravimetric Analyzer (TGA/DSC)	Co, Li	[38]
9	Atomic Emission Spectrometer	Co, Ni, Li	[109]
10	Atomic Adsorption Spectrometry (AAS)	Co, Mn, Ni, Al, Fe, Cu, Li, Na, Gr	[31, 94, 102, 110]
11	Flame Atomic Adsorption Spectroscopy (FAAS)	Cu, Co, Mn, Fe, Ni, Li	[105]
12	UV-visible (UV-vis) spectroscopy	Li, Co, Ni	[111]

The leaching kinetics was quick despite the inadequate yields of Li and Co (60–85% for Li and 50–75% for Co) in the HCl media used for the tests. Due to the variability of the raw materials, some elements, like copper and aluminium, were enhanced into larger portions, which presented difficulties for the laboratory-scale studies. Viscous slurry conditions were created throughout the leaching process by the plastics and graphite found in the LIB waste. A sizable amount of solids (about 40% of the input materials) remained undissolved in the leach residue [105]. This proves that the proper selection of the correct leaching media is crucial.

To avoid inadequate leaching, additional reactants or additional processes can also be included. For example, when vacuum pyrolysis and acid leaching were combined, the cobalt leaching efficiency increased by adding H_2O_2 (hydrogen peroxide) to the H_2SO_4 utilized for leaching. Under ideal circumstances, a 99% leaching efficiency for Li and Co was achieved [114].

However, using inorganic acids is linked to many emissions, including the generation of waste streams and wastewater, as well as gas releases (SO_3, Cl_2, or NOx, according to the acid used) [47, 109]. Organic acids are typically more benign to the environment compared to traditional inorganic agents because they don't produce gaseous emissions [47, 109, 115]. Numerous organic acids have been researched for recovering wasted batteries; citric and malic acids have received the most attention [47, 115].

The procedure of leaching used $LiCoO_2$ materials in a DL-malic acid and H_2O_2 system was substantially improved by a one-step revitalization process and ultrasonic improved leaching of $LiCoO_2$ materials. Leaching efficiency of 98.13% for lithium and 98.86% for cobalt were demonstrated under optimal circumstances of 1.5 mol/L DL-malic acid with 3 vol% H_2O_2, 4 g/L solid/liquid ratio, 95 W ultrasonic power, 80°C temperature, and 25 minutes leaching time [39].

Using mild tartaric acid as a leaching agent, as well as a precipitating reagent, an ecologically benign approach was described for recovering high-value metals (Co and Li) derived from common waste cathode materials ($LiCoO_2$) of discarded LIBs. More than 97% Co and 98% Li were leached and precipitated under optimal leaching circumstances: acid concentration of 0.6 mol/L, duration of retention of 30 minutes, reductant dose of 3 vol% H_2O_2, density of pulp of 30 mL/g, and temperature of reaction of 80°C [38]. Mild tartaric acid was used in another experiment to recover precious metals (Co and Li) from standard discarded cathode components ($LiCoO_2$) of used LIBs. Here, a leaching efficiency of greater than

99% for Co, Mn, and Li was achieved in the optimal leaching circumstances, including a density of pulp of 20% (w/v), HCl 1.75 M, temperature of 50°C, and duration of 2 hours [116].

Weak acids, however, had to be combined with a reducing agent because they were discovered to be insufficiently strong to dissolve the oxides [47, 115]. Therefore, attempts were made to use stronger organic acids as leaching agents.

It was looked into if oxalic acid, which is the strongest organic acid, might be used as a leaching agent to recover lithium selectively early on. The leachate included only soluble lithium simple oxalate and aluminum complex oxalates that could be selectively dissolved under ideal conditions; the remaining transition metals remained in the remnants from leaching. This might greatly enhance the impurity removal process because it is difficult to separate aluminum from the black mass with the conventional sodium hydroxide method [47].

Recovering cobalt, nickel, and lithium from used LIBs in an efficient manner was demonstrated by a study concentrating on low leaching temperatures in conjunction with organic acids (citric, oxalic, and formic acid) [109].

There have been instances where the hydrometallurgical process has been combined with vacuum pyrolysis. In experiments, $LiCoO_2$ was leached and precipitated using oxalate as a leaching agent and precipitant. The efficiency of leaching $LiCoO_2$ rose along with the oxalate content and length and temperature of the reaction. Under ideal circumstances, the maximum reaction efficiency of 98% of $LiCoO_2$ was achieved, enabling the recovery and separation of Li and Co from spent LIBs [117].

The newly developed low-cost, secure, and eco-friendly solvents are called deep eutectic solvents (DES). They have an amazing capacity to dissolve metal oxides and are considered ecologically benign substitutes for conventional acid-leaching. A closed-loop procedure aimed to recover the metals through antisolvent crystallization after leaching $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ (NMC) using a DES (choline chloride and L-(+)-tartaric acid). Evaluations were conducted on five organic antisolvents: 2-propanol, acetone, ethanol, methanol, and ethylene glycol. Among them, ethanol demonstrated the highest metal recovery efficiency of Co (>98.5%), Ni (>98.5%), Mn (>96.5%), and Li (70%). Compared to ethanol, ethylene glycol caused recovery yields of transition metals to drop by up to 8%, yet it allowed for the efficient separation of Li. However, acetone and 2-propanol were found to be unsuitable as antisolvents because of their immiscibility with the DES leachates [97].

The technique of ion separation and inorganic acid leaching has a high leaching efficiency and produces highly pure metal products. Nonetheless, acid consumption must be managed. More optimization is needed to determine the ideal parameter throughout the leaching process [34].

Apart from the acid leaching method, research has also been done on an alkaline system using ammonia [72]. Through the addition of sodium sulfite as a reductant and ammonia-ammonium sulfate as the leaching solution, the first-step leaching solution yielded a total selectivity of over 98.6% for Ni, Co, and Li from the cathode scrap of spent lithium-ion batteries and 1.36% for Mn [118]. Similar research was conducted using ammonia, ammonium carbonate, and ammonium sulfite-based ammoniacal leaching agents to examine the way treated cathode active materials leached Ni, Mn, Co, Al, and Cu removed from an industry-standard LIB pack in electric hybrid cars. Leaching kinetics were improved by ammonium sulfite as a reductant, especially when ammoniacal leaching of nickel and copper was involved. The pH could be buffered using ammonium carbonate. While Mn and Al were rarely leached and Ni had a moderate leaching efficiency, Co and Cu could be entirely leached off [119]. To remove aluminum selectively, the material for the active cathode powder was initially leached using a 5% by-weight NaOH solution. After that, the residues were again leached using an H_2SO_4 solution. Under ideal circumstances, lithium and cobalt had respective leaching efficiencies of 96% and 95% [120].

It has always been a debatable point whether recycled metals would be of appropriate quality to be used in manufacturing new LIBs. Some studies have successfully obtained usable quality materials by recycling LIB wastes. Scraps of copper, aluminum, and steel that made up around 30% of the initial weight of the LIB trash were recovered by

basic treatments in a closed-loop hydrometallurgical process. After the black mass underwent hydrometallurgical treatment, additional waste fractions were recovered, comprising 1%–3% of nickel hydroxide/oxide, 10%–12% of cobalt products, 1%–16% of graphite, and 1%–2% of lithium carbonate. The procedure yields graphite, $CoC_2O_4{\cdot}2H_2O$, and Li_2CO_3, all of which largely meet market criteria. It is also possible to obtain other products meeting commercial standards (Na_2SO_4) and added-value compounds (Mn–Co magnetic nano ferrites) [94].

Efforts have also been made to introduce techniques where the usage of acids could be reduced, as they produce huge volumes of wastewater with high concentrations of corrosive substances. One such method was the comprehensive leaching of important metals (Li, Co, Mn, and Ni) from recycled lithium-ion batteries, devised and tested with the help of electrochemical assistance. The suggested method uses electrons as a green reagent to replace chemicals in a hydrometallurgical leaching process. By using this method, modest amounts of Fe^{2+} can be used to regenerate and replace hydrogen peroxide as a reducing agent. It reduces the need for acid because H^+ can be produced electrochemically. It was possible to attain above 96% leaching efficiency for Mn, Li, Co, and Ni, in addition to electrowon Cu and graphitic carbon by-products. Furthermore, a lower pH for the leachate indicates further reductions in chemical usage (pH correction) for the downstream metal extraction process [121].

As more demanding applications require higher levels of electrochemical performance, longevity, and safety, it is anticipated that a sizable percentage of future LIBs will be made up of all-solid-state batteries (ASSBs), which use a solid-state electrolyte (SSE) rather than a traditional non-aqueous liquid electrolyte. However, recycling considerations for ASSBs are not fully investigated. ASSBs and traditional LIBs were compared regarding recycling, and the feasibility of several recycling methods was examined using hydrometallurgical and direct methods. According to a study, well-designed equipment can significantly raise LIB (here, NMC) recycling rates above the existing 5% levels [122]. Such methods are distinguished by their lower water usage and avoidance of the need for organic solvent extraction. More research is necessary to compare this procedure to other recycling methods while taking into account its efficiency as well as its effects on the environment and the economy [97].

Biohydrometallurgy is a field of biotechnology that uses biological systems and organisms to efficiently recover metal from solid compounds by producing components that may be extracted [123]. The environmentally acceptable ways of microbial engagement for recovering metals from waste LIBs have made it a desirable technique. Among the organisms participating in the recycling of metal from LIB are Lysinibacillus, Micrococcus, Sporosarcina, Empedobacter, Barrientosiimonas, Paenibacillus, Bacillus, and Acidithiobacillus [124]. The three bioremediation techniques most frequently used are bioleaching, biosorption, and bioaccumulation [125].

Aspergillus niger was used in a laboratory setting to investigate the one-step, two-step, and bioleaching of wasted medium of used mobile phone batteries. The outcome showed that spent medium bioleaching produced a larger overall recovery than other techniques; one hundred percent copper, 95% lithium, 70% manganese, 65% aluminium, 45% cobalt, and 38% nickel were leached. In one-step bioleaching, the leaching of 100% lithium, 58% aluminium, 11% copper, and 8% manganese was observed, while nickel and cobalt leached very little. One hundred percent lithium, 61% aluminium, 10% manganese, 6% copper, and 1% cobalt were leached in two steps of bioleaching [108].

Under industrially relevant conditions, a bioleaching process capable of recovering essential materials from end-of-life lithium-ion batteries (EOL-LIBs) was developed. The cathode-containing powder, or black mass, made from trash LIBs was leached utilizing a biolixiviant that the Gluconobacter oxydans bacteria manufactured from corn stover. Fe(II) was added as a reducing agent to help the metals dissolve. Recovery of up to 86% of cobalt, 100% of Li, 100% of Mn, and 84% of nickel from LIB black mass was accomplished with the optimal leaching temperature of 55°C, pulp density of 2.5%, gluconic acid concentration of 75 mM, and leaching time of 30 hours. According to

life cycle assessment (LCA), bioleaching of used LIBs has the potential to be both more cost-effective and ecologically friendly compared to alternative hydrometallurgical recovery methods [126].

The hydrometallurgical process typically has two disadvantages: it uses a lot of chemical reagents, and it is hard to do away with contaminants from the recovered product [48]. The spent LIBs' active electrode materials recycling procedure has sparked considerable research, but the separation of metals (copper, aluminium, iron, and nickel), which are mostly loose particles in coarse sizes and possess a substantial financial value, has not received as much attention [51].

Employing a range of precipitants, including dimethylglyoxime (DMG), $(NH_4)_2C_2O_4$, and Na_3PO_4, the nickel, cobalt, and lithium ions present in the leachate derived from commercial $LiCoO_2$/$LiNiO_2$ cathode substances using citric acid and hydrothermal leaching were precipitated and separated. While the metals were being separated, variables like pH level, amount of precipitant, and reaction temperature were optimized. Nickel, cobalt, and lithium recovery rates were 97.2, 96.1, and 94.1%, respectively, and their respective precipitates had Ni, Co, and Li purities of 96.3, 96.2, and 99.9%. While hydrothermal leaching requires pressure-resistant reactors and is carried out at greater temperatures than standard leaching using a reductant (such as H_2O_2), it provides better industrial application, lowers the amount of chemicals consumed like reductants, and accelerates the reaction pace [111].

Using a combination of pretreatment, mechanical treatment, and hydrometallurgical process, the $LiFePO_4$ cathode was synthesized from spent LIB (LFP battery) by precipitation. Mild organic acids, like p-toluene sulfonic acid (TSA) and methyl sulfonic acid (MSA), were used for leaching at room temperature. High extraction efficiency (95%) of lithium and iron from the LIB black mass was attained [104].

The removal of 62% of Al and 75% of Cu was achieved through density separation in another study that combined mechanical and hydrometallurgical processes. Iron (99.6%), copper (81.3%), and aluminium (80.5%) could be eliminated through neutralization, and nickel and copper carbonate precipitation was then used to extract Ni (97.1%), Co (97.2%), and Mn (97.3%). In the end, precipitation generated Li_2CO_3 with a 50% yield and a comparatively high purity of 95% [105]. Despite such wide application of the hydrometallurgical method, the lack of versatility in raw material disposal remains a significant difficulty.[72] Also, owing to possible environmental risks, it is necessary to recover the electrolyte and the materials used in the anode from used LIBs besides the cathode active elements [83].

The metallic ions moving selectively transitioning from the watery to the organic phase, facilitated by complexation agents (extractants), is known as solvent extraction. A strong acid is used to "strip" the metal from the organic phase. The metal is then physically recovered from the aqueous solution by precipitation or reduction, and the extractant is recycled [3].

In a continuous procedure for recovering cobalt from used cell phone batteries, the materials were first disassembled, crushed, and categorized. Following an investigation on leaching using hydrogen peroxide and sulfuric acid, cobalt was selectively separated using liquid-liquid extraction. By evaluating the effects of the concentration of the extractant, pH, and the length of contact on the extraction percentage, the ideal extraction circumstances were identified. A comparable investigation was conducted for the stripping conditions, and cobalt electroplating was carried out as a final step. When the extractant concentration was between 0.4 mol/L and 0.5 mol/L and the pH was between 6 and 7, the extraction yields were the greatest, ranging from 97-99% [31].

On using HCl as a leaching media, impurities were also attempted to be eliminated by neutralizing with NaOH at various temperatures after mechanical separation. Under ideal circumstances, the majority of iron (99.6%), copper (81.3%), and aluminium (80.5%) could conceivably be taken out (pH = 5, T = 50°C). Following neutralization, precipitation of nickel and cobalt carbonate (at T = 50°C and pH = 8) was employed to extract Ni (97.1%), Co (97.2%), and Mn (97.3%) [105].

Cyanex 272 is an efficient method for the purpose of recovering specific metals from waste cathodic active material, which can help reduce environmental pollution, recycle trash, and preserve natural resources. Temperature, pulp density, the concentration of leaching agent (H_2SO_4), and reductant (H_2O_2) concentration were considered while leaching the waste. Under ideal circumstances, 94% of lithium and 93% of cobalt were leached. Finally, cobalt-loaded organic was stripped with H_2SO_4, giving a 99.99% pure cobalt sulfate solution [127].

After purification, approximately 98.7% of nickel could be precipitated and recovered under ideal circumstances using dimethylglyoxime reagent by selective precipitation and solvent extraction from the leaching liquid of wasted LIBs. After that, the precipitate was dissolved in 1 mol/L hydrochloric acid. White dimethylglyoxime powders that have been regenerated could be utilized as a precipitant again after filtering. Consequently, Co-D2EHPA could extract manganese up to 97.1%. Using ammonium oxalate and hot-saturated sodium carbonate, cobalt and lithium were finally selectively precipitated, recovering approximately 98.2% as CoC_2O_4 $2H_2O$ and 81% as Li_2CO_3 [128].

Much unseen, the application of adsorption has also been tested on LIB recycling. Solvent-impregnated resins (SIRs) were investigated as a recycling method for recovering precious metals (Cu^{2+}, Co^{2+}, and Li^+) selectively from the synthetic leachate of waste LIBs to overcome the drawbacks of the technique of solvent extraction, which included the strong phase mixing, the need for significant volumes of organic solvent, the formation of a third phase, and the organic solvent being lost and entering the aqueous phase. The recovery was accomplished by one chemical precipitation (for Fe^{3+} and Al^{3+}) and four chromatic operations (for Cu^{2+} and Co^{2+}) [129]. In another experiment investigating the efficiency of adsorption for recycling of spent LIBs, it was found that compared to multivalent ions like Mg^{2+}, Li^+ in states of low valence exhibited substantially poorer adsorption (and energy absorption) through graphene. However, thermally reduced graphene oxide (rGO) membranes were found to be capable of efficiently lowering the high mass ratio of a combination Li^+/Mg^{2+} solution, which was 500:1, to 0.7:1 in the Mg^{2+}/Li^+ separation process [130].

The solvent extraction procedure has a greater environmental impact because it handles wastewater and does not recycle lithium [131].

The hydrometallurgical method for recycling spent LIBs has gained a lot of popularity recently. Blended cathode materials (BCMs) are widely used in EVs. Li and Fe were recovered from BCMs of used $LiFePO_4$ and $LiNi_xCo_yMn_{1-x-y}O_2$ batteries. Alkali leaching removed 87% of the Al, and selective leaching with H_2SO_4 and H_2O_2 separated 91.65% Li, 72.08% Ni, 64.6% Co, and 71.66% Mn. After being leached, 98.38% of the iron was recovered as $FePO_4{\cdot}2H_2O$, which was precipitated to provide 99.5% pure iron [132]. T. Tawonezvi et al. combined potentiostatic and hydrometallurgical techniques to recover precious Ni-Co alloys made from synthetic solutions of nickel, cobalt, manganese, and lithium sulfate similar to the NMC532 ratio. It proved to be an economical substitute for the power-intensive, money-intensive, and material-intensive intermediate purification techniques for hydrometallurgy like ion exchange, solvent extraction, and selective precipitation. 98.2% pure $Ni_{0.65}Co_{0.35}$ was recovered by optimizing parameters such as applied voltage, temperature, pH, Co, and Ni content of the NaH_2PO_4 buffer, Na_2SO_4, and cathode rotation speed [133]. Reducing agents make the waste streams generated in the leaching process more corrosive. NaOH was used in a hydrometallurgical process to recover wasted $LiFePO_4$ cathodes due to its selectivity for Li and ability to function simultaneously as an oxidant and a leaching agent. Lithium and phosphorus leaching efficiencies of 98.2% and 99.9%, respectively, were achieved, and 100% of the iron was changed to Fe_3O_4. Moreover, Na^+ might intercalate the $LiFePO_4$ structure. Compared to traditional techniques, this approach was described as more affordable, effective, and ecologically friendly [134]. Interestingly, it doesn't seem that leaching or high temperatures alone produce very satisfying results. While the ammonia complexation environment created by the breakdown of cysteine (Cys) allowed for effective transition metal leaching without the need for further additions, the lithium in $LiNi_xCo_yMn_zO_2$ (NCM) was able to be reduced with zinc powder to

produce soluble LiOH, giving 92% and greater than 97% leaching effectiveness for Li and Ni/Co/Mn, respectively [135].

Another study attempted to create the hydrometallurgical recycling method for NCA cylindrical batteries because there aren't many cost-effective recycling studies on NCA batteries, which account for 8% of the market share. Three acids were examined for their potential to leach: citric acid, H_2SO_4, and H_3PO_4. Citric acid came out as the most profitable option. Al foils eliminated the requirement of reducing agents, thus reducing the leaching cost. It was possible to recover almost 92% of Li, 80% of Ni, and 85% of Co. However, the separation of solids and liquids remained a problem [136].

Metals have also been removed using low-cost, reusable bio-sorbents. For instance, orange peels and sugarcane bagasse removed 96.71% and 94.74% of Mn(II), respectively, at optimum contact periods of 120 minutes for orange peels and 30 minutes for sugarcane bagasse from lab-synthesized and actual effluents [137].

Compared with the pyrometallurgical process, hydrometallurgy takes less energy, produces minimum amounts of toxic gases, and exhibits better efficiency for metal recovery. The selection of the leaching media depends on the elements present in the black mass and also on the targeted metal. However, the inconsistency in the composition of batteries makes this selection very difficult. Inorganic leaching acids that generate harmful gaseous emissions and corrosive wastewater streams can be replaced by organic acids. Weak organic acids, however, need the addition of reducing agents to achieve effective leaching, whereas strong organic acids alone are sufficient. Owing to the huge consumption of chemicals, handling the waste stream from hydrometallurgical processes becomes very difficult. Therefore, optimization of the process must be done to manage the acid consumption. To do away with the risks associated with acid leaching, alkali leaching must be uplifted, which has shown improved leaching kinetics in the past. Deep eutectic solvents have emerged as cheaper and environmentally benign substitutes for conventional leaching. Bioleaching, biosorption, and bioaccumulation also leach the black mass without creating any environmental nuisance.

New techniques must be developed to combine hydrometallurgy with other physical methods. For instance, the need for chemical reagents can be eliminated due to the generation of H^+ ions when hydrometallurgy is electrochemically assisted. Density separation before hydrometallurgy can make targeting particular metals easier. Different adsorbents may be suitable for adsorbing ions with different valencies, thus eliminating them from the solution and leaving others for further processes. All-solid-state batteries (ASSBs) should also be encouraged to avoid the use of traditional liquid electrolytes.

Recycling metals from used LIBs remains a challenge despite the existing and widely used pyrometallurgical and hydrometallurgical methods because these two systems cannot be considered green methods because they involve the use of harmful chemicals and lead to the generation of toxic effluents in the form of gas and liquid. These drawbacks call for other methods that can go in harmony with the principles of carbon neutrality and sustainability, achieving a circular economy [138].

7.3 Direct recycling

Direct regeneration is a recycling strategy that is preferred due to its favorable effects on the environment and economy [98]. The widely used pyrometallurgical and hydrometallurgical recycling techniques take up huge amounts of energy, need hazardous chemicals to bring about the desired reactions, and discharge harmful materials. Thus, these processes become incompatible with the concept of circular economy. Direct regeneration is a non-destructive and sustainable repair technique that uses non-toxic reagents and maximizes the merits of reclaimed LIB electrodes [139]. Using the least amount of energy and causing the least amount of disturbance to the environment, the direct recycling process transforms materials from end-of-life (EoL) cathodes into materials suitable for batteries [96].

Four LIB kinds (NCA, NCM622, NCM811, and NCM111) were evaluated using three distinct approaches to recycling, i.e., pyrotechnical recycling (PR), hydrometallurgical recycling (HR) and direct physical recycling (DPR), to evaluate

these techniques' respective efficiencies. This study demonstrated that DPR has far greater environmental and economic benefits than the other two recycling technologies, even though it is still in the early stages of development [53]. The prices, energy inputs, and greenhouse gas emissions related to the production and recycling of lithium-ion batteries making use of three typical cathode chemistries—lithium nickel manganese cobalt oxide (NMC-622), lithium nickel cobalt aluminum oxide, and lithium iron phosphate—were investigated by models of process-based costs and an attributional life-cycle analysis. It was observed that direct cathode recycling can lower emissions and be economically competitive [140].

Technologies for cathode-to-cathode, mechanical, electrochemical, and cathode-healing are examples of direct recycling techniques. Through pyro- and hydrometallurgical recycling methods, lithium recovery from end-of-life LIBs requires multiple stages of refining, including the consumption of a significant amount of energy and chemicals. Attempts have been made to solve this problem. For example, a workable method for creating distinct metal concentrates was the amalgamation of magnetic separation and flotation. The findings demonstrated that plastics were easily removed from the combination of metals using the standard flotation technique when methyl isobutyl carbinol (MIBC) was present. Upon completion of the copper flotation circuit, a 53.5% copper concentrate was extracted following plastic flotation. To distinguish between diamagnetic aluminium particles and ferromagnetic metals (Fe and Ni), tailings from copper flotation were subjected to dry magnetic separation; around 99% recovery was obtained [51].

Density-based separation of 62% for Al and 75% for Cu was achievable in an experiment where the materials were mechanically processed to examine the suitability of sieving and density separation; nevertheless, other valuable components stayed attached to these fractions. The Cu-rich overflow contained a significant amount of Co (> 20%), which suggested the existence of polymeric binders. Consequently, further steps may be needed after mechanical separation to produce a substance that has a greater capacity to separate Cu and Co-rich fractions [105].

Among the most effective methods for recovering lithium is the electrochemical method, which has a wide range of benefits, including low energy usage, quick reaction times, and excellent lithium selectivity [103]. Li extraction was achieved by aqueous electrochemical delithiation of $LiMn_2O_4$, $LiCoO_2$, and $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$, yielding extraction efficiencies of 95%, 97%, and 99%, respectively. 80%, 95%, and 99% associated faradaic efficiencies were achieved, with negligible co-extraction of cobalt, nickel, and manganese. This process ought to be seen in the role of a pretreatment stage in the recycling of the black mass of LIBs, which contains Cu, Fe, Al, and graphite. By using this method, high-purity lithium may be selectively extracted and recovered without having to undergo the laborious process of separating Li from the residual solution that is left over after the hydrometallurgical process [106].

A unique method of relithiation by electrochemical means in an aqueous medium, succeeded by thermal treatment of recycled $LiCoO_2$ substances, showing similar morphology, crystal composition, and electrochemical efficiency to the $LiCoO_2$ sold commercially [96]. The spent $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ cathode material was reduced to Li_2CO_3, NiO, Co_3O_4, Mn_2O_3, and MnO_2 by a unique and ecologically friendly process of combined heat reduction and electrochemical leaching. Tests for electrochemical leaching reveal that 100% Li, 90.59% Ni, 90.53% Co, and 66.40% Mn can be leached. Electrochemical leaching for roasted products is a cost-effective and efficient method that offers several benefits, such as high leaching efficiency, no need for a reducing agent, reduced acid usage, and complete utilization of depleted resources [141].

An electrochemical membrane reactor was created with the capability to electroplate copper and electrochemically precipitate aluminum and iron from artificially produced leachates of expended LIBs by merely using electrical energy, water, and air and reducing impurities to <1 ppm. The leachate thus obtained contains 95.4% of the Co, 99.5% of the Ni, and 99.14% of Mn from the initial solution of leachate and is directly applicable to the synthesis of cathode predecessors. This method

requires much less chemical consumption, doesn't introduce any extra impurity or greenhouse gases (GHG), and also generates a valuable by-product in the form of hydro sulphate (H_2SO_4), making it a cost-effective process [102].

Dielectrophoresis (DEP) is an electrokinetic method of manipulating particles that enables the selective separation of particles according to their substance, size, and shape. It separated graphite and lithium iron phosphate (LFP), two typical materials for LIB cathode and anode. Where LFP demonstrated a low separation efficiency of 1–15% from a blend of LFP and graphite, graphite exhibited a high effectiveness of separation of 70–100% [110].

Lithium manganese oxide (LMO) and activated carbon (AC) electrodes were used in electrochemical sorption to recover lithium from real industrial-scale LIB leachate successfully. In the LIB leachate, the LMO electrode demonstrated a high degree of selectivity for lithium relative to additional cations that coexist, including nickel, cobalt, manganese, calcium, aluminium, copper, iron, and magnesium. It was found that an extended reaction time and an increased current boost lithium recovery's capacity, purity, and specificity [103].

Because the present processes either damage the cathode's structure or need an excessive amount of toxic and costly chemicals, they have certain limits for direct regeneration when it comes to separating the active cathode components from the aluminum foil. As a result, a thorough analysis was conducted on the properties and development of NCM-based cathodes following cycling, separation, and regeneration. An incredibly straightforward, effective, and environmentally friendly process was created to effectively separate the cathode elements off the aluminium foil, directed by the growing amount of lithium molecules that remain on the particle surface. Secondary pollution won't happen because this method does not use organic solvents. Additionally, the recycled aluminum foil stayed intact and could be employed immediately in the industry [98].

A straightforward, direct recycling method was successfully used to demonstrate the full regeneration of deteriorated LMO cathodes. The one-step hydrothermal treatment in diluted Li-containing solutions allowed for the flawless reconstruction of the appropriate stoichiometry and phase purity, which resulted in the increased capacity of regenerated LMO particles, prolonged cycling stability, and performance at a rapid pace. According to lifecycle analysis, the work offered a simple but effective method of re-functionalizing high-performance LMO cathodes, with clear advantages over conventional pyrometallurgical and hydrometallurgical processes in terms of both the environment and economy [101].

Relithiating spent materials of cathodes usually demands high temperature as well as high pressure. This issue is an obstacle to the industrial application of the direct regeneration method [142]. Direct replenishment of depleted $LiFePO_4$ (LFP) electrodes was demonstrated by coupling quick post-annealing with a low-temperature aqueous solution relithiation based on defect-targeted healing. This process depicted a considerable decrease in energy consumption and greenhouse gas (GHG) emissions, as well as the revival of the formation, composition, and electrochemical activity of the regenerated LFP to levels equivalent to the pristine LFP [107].

Using the Taguchi Design of Experiment (DoE), the efficiency with which cathode materials can be recovered in direct recycling of end-of-life (EoL) LIBs was methodically investigated. A statistical regression model was created to forecast the yield and direct the choice of parameters. The study determined that the three crucial control elements for the material recovery yield through the extraction of organic solvents were the sonication period, sheet dimensions, and the ratio of solid to liquid weight. The mathematical relationship between the yield and control parameters was successfully established, and confirmation tests demonstrated its accuracy [143].

A thorough examination of the cathode material's failure mechanism in spent LIBs revealed that the spent NCM523 material had issues with structural transformation, surface interface failure, and the dissolution of lithium and transition metals, all of which significantly worsened the electrochemical performance of the material. Through granulation, ion doping, heat treatment, and metal ion supplementation, the NCM523 material was directly renewed. During the recovery process,

PO_4^{3}-polyanions were added to the NCM material to be regenerated, exhibiting superior electrochemical performance. This work expedites the commercialization of extended LIBs recycling by offering a fresh strategy for the recuperation of different lithium nickel cobalt manganese oxide (e.g., $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$, $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$, $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$, and $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$) [144].

An innovative, environmentally friendly technique was presented in order to recycle used single-crystal $LiNi_xCo_yMn_{1-x-y}O_2$ (NCM) components, in which the direct calcination procedure did not induce any new chemicals. This technique would make it possible to significantly enhance the electrochemical performance of discarded cathode materials and reuse the Li contents in a highly efficient manner [145].

Using the aluminium contamination coming from the corresponding current collector to provide the necessary transition metals for both structural restoration and elemental compensation at the same time, a two-step procedure was created for the direct recycling of deteriorated NCM components, which was capable of regenerating high-quality NCM material. The exceptional electrochemical performance of this recycled NCM substance with controlled aluminium doping was even on par with that of new materials [146].

An effective and environmentally friendly technique for regenerating a deteriorated $LiFePO_4$ cathode was developed. Using hydrogen peroxide as a leaching reagent, the sheets of cathode were straight divided into aluminium foil, superior purity $FePO_4$, and a lithium-containing mixture. With limited Al and Fe, the leaching efficiency of Li surpassed 96.3%, and the recovered $FePO_4$ yields regenerated $LiFePO_4$ with exceptional rate capacity and steady cycling over time. The amount of chemicals used and the number of processing stages were both greatly reduced by this study, which is beneficial for the long-term industrial growth of used LIBs [147].

The recent literature trend indicates an inclination to use the non-destructive direct recycling process. For instance, by annealing wasted graphite in CO_2, CO_2 roasting was utilized to restore the graphite structure. This changed the metal contaminants into carbonates that were soluble in water and removable via aqueous washing. More efficient than N_2 roasting in removing adulteration from graphite, CO_2 roasting enhanced the rate functionality of regenerated graphite. This environment-friendly process did not use acid for leaching, thus solving the problem of acid waste disposal [148]. In contrast with the destructive recycling methods used currently, which destroy the three-dimensional structure of the cathode materials by heat treatment or liquid extraction, the Flash Joule Heating (FJH) method, in combination with magnetic separation, reinstated new cathodes from waste ones in milliseconds and with 98% metal recovery. The method exhibited better environmental and economic sustainability [149].

In another scenario, after discharging, the waste LFP battery was dismantled to extract the cathode material. Following the elimination of impurities, Li^+ additions, carbon nanotube (CNT) doping, covering of carbon, and thermal processing, the waste LFP material was restored. High temperature improved the morphology of waste LFP via Li^+ supplementation; whereas carbon-coating made the structure more stable. The conductivity of LFP and its ionic diffusion coefficient were significantly increased by the 3D conductive networks made of CNTs [95]. Using an amalgamation of sonication, screening, and multiple-step flotation (with n-dodecane acting as collector and methyl isobutyl carbinol, MIBC, as frother), it was possible to successfully isolate the black mass from LIBs into comparatively pure LFP and carbonaceous material components. Higher purity levels for the carbonaceous and LFP material fractions were obtained using double flotation. Graphite and LFP showed recovery rates of 80% and 75%, respectively [150]. For recycling, the cathode of a LIB is preferred, and recycling of anode materials is neglected because of the poor value-added results in resource waste and environmental pollution. The anode materials were reused through a direct regeneration process that used discarded graphite as the core and asphalt as the coated carbon supply. Liquid phase impregnation was used to create asphalt pyrolyzed graphite composites, and high temperatures restructured the wasted graphite to increase the degree of graphitization and interlayer lattice spacing. The surface morphology was smoother after asphalt coating.

When utilized as anode material, the regenerated graphite demonstrated acceptable electrochemical performance [151].

Like any other recycling method, direct recycling also faces certain hurdles. The problem facing the environmentally acceptable and economically viable direct recycling approach is the unequal allocation of components and particle dimensions in used cathode components. In a depleted $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ cathode, chlorination was employed as a pre-treatment to remove Li and improve particle size uniformity selectively. Repurposed monocrystalline $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ (NCM811) was successfully synthesized by a multi-step aging method that enhanced the primary particle size and reduced impurity production [152].

The non-destructive and sustainable method of direct regeneration is comprehensively preferred over pyrometallurgy and hydrometallurgy as it consumes much less energy, makes no use of harmful chemicals, does not discharge toxic substances, reduces the emission of greenhouse gases produced during other recycling processes, and is economical and environmentally friendly. The direct regeneration method uses physical and mechanical processes such as cathode-to-cathode, mechanical, electrochemical, and cathode-healing to transform used battery cathode materials into materials suitable for producing new batteries without changing their chemical compositions. The electrochemical method used for direct recycling shows high-purity lithium selectivity without the need to separate it from the superfluous solution as in the hydrometallurgical process. Also, the electrochemical method followed by heat treatment recovers commercially competent $LiCoO_2$. Direct recycling combined with a step of hydrothermal treatment enhances the capacity of regenerated battery materials, gives longer cycling stability, and makes the battery perform at a faster rate.

New direct recycling techniques show promise, but they also have drawbacks, including improper battery labelling, the logistical difficulties of collecting dead batteries effectively, and component separation [8]. After physical processes like sieving and density separation, valuable metals remain adhered to each other, necessitating further steps to obtain metals of greater purity. Electrochemical separation has also been seen to be slightly biased towards lithium recovery over other metal ions. Moreover, the direct recycling process is in its early stages of development, and hence, the available literature does not give complete knowledge about all its loopholes. More and more implementation of this method will eventually bring forth other significant drawbacks, if any.

No technology for recycling batteries is perfect at present, and some obstacles are there that need to be addressed. Potential improvements in the process can be achieved by governments, businesses, and academia working together [11]. Table 7 compares the three methods discussed above [153].

8. Comparative analysis of the existing recycling and recovery methods and scope of future research

The systematic literature review of the various recycling methods for spent lithium-ion batteries indicates the following. The pyro-metallurgical method, though, is an established system having industrial implementations; it has the associated drawbacks of toxic gas emissions, excessive energy consumption, and selective metal recovery. This recycling method utilizes a high-temperature system that liquefies and breaks up different elements of the battery, which can contribute significantly to greenhouse gas emissions. More research is required on enhancing the lithium and other precious metal recovery by pyrometallurgy using combination or hybrid methods and process optimization to minimize the environmental impact of pyro-metallurgy.

Hydrometallurgical recycling involves usage of chemicals for dissolving and recovering precious metals from waste battery parts. Hydrometallurgical recycling is a better option than pyro-metallurgical processes from the perspective of environmental impacts and can further be improved by optimization of chemical consumption that yields higher efficiency and by practicing recycling and reuse of solvent. However, it has still not found industrial applications for many battery chemistries. The impurities developed and the associated scaling up are

difficult to control in large-scale applications, indicating the future scope of research in this area. Direct recycling or direct recovery methods are emerging technologies that can directly reuse the battery elements by minimizing the use of hazardous chemicals, as in the case of hydrometallurgy. It is energy efficient and provides selective recovery of highly pure precious metals. Similar to the hydrometallurgical system, this is also in the early stages and comes with the challenges of impurity generation and scaling up.

There is research on comprehensive life cycle assessment to assess their general environmental and economic impacts, which indicates creating closed-loop systems of recycling that generate minimum waste and boost the reuse of materials. These studies also show the necessity of designing the batteries while considering recyclability as a primary concern, which can be achieved by using easily separable elements and non-hazardous substances. Continuous research and development are needed for new techniques, like membrane processes for separation, bio-recyclers, and solvent extraction, to refine the efficiency of battery recycling and make it more sustainable.

Table 7. Advantages, drawbacks, and future scope of different treatment processes and metal recovery methods on LIB waste.

Sr. no.	Method	Advantages	Drawbacks	Scope for improvement
1	Pyrotechnical recycling	1. Simple 2. Highly productive 3. Applicable at a large scale	1. High energy consumption 2. Serious air contamination 3. Additional steps needed for lithium recovery	1. Pretreatment of electrolyte 2. Development of a green efficient purification system
2	Hydrometallurgical recycling	1. Energy efficient 2. Cheap 3. Simple 4. Less polluting	1. In laboratory stage 2. Difficult to control impurities	1. Large scale applicability 2. Product quality improving
3	Direct recycling	1. Products of high purity 2. Energy efficient 3. Capable of selective extraction of valuable metals 4. High recovery rate 5. Several high-value-added products	1. In laboratory stage 2. Difficult to control impurities	1. Large scale applicability 2. Product quality improving

9. Integration with circular economy

Amalgamating recycling of LIBs into the framework of circular economy requires minimization of waste generation and maximizing the utilization of available resources. This implies developing easily recyclable batteries, setting up proper collection systems and storage facilities, and creating transformational techniques to recover critical metals and reusing these metals in fresh batteries and other items. Adopting a circular economy minimizes the dependability on virgin minerals and, thus, reduces the related environmental hazards by creating a more sturdy and sustainable battery production and recycling system.

Electrodialysis is an efficient method that can achieve the target of circular hydrometallurgy for recovering critical metals. However, electrodialysis faces challenges such as excessive utilization of energy and process optimization [154]. Efforts have also been made to replace virgin metals with other innovative materials. For instance, polyethylene terephthalate (PET) waste can be used in

manufacturing lithium-ion batteries. PET has exceptionally good mechanical properties and is resistant to most of the chemicals. This makes them an excellent option for a raw material for LIBs [155]. Integrating artificial intelligence (AI) into the circular economy also enhances the recycling process by supporting the principles of circular economy. Involvement of industries, upgradation of existent technologies and development of more advanced ones, and cloud-based tracking with analysis giving battery health evaluation independent of vendors are encouraged [156]. Another challenge faced by the current recycling technologies is pollution due to secondary heavy metals and volatile organic pollutants since the present systems of recycling primarily focus on the recovery of high-value metals and are barely concerned about reclaiming elements of lower value like graphite, separators, and electrolytes. These processes are also very lengthy and expensive [157]. Circular economy supports the development of methods that aim for zero-waste generation and recovery of all the components of LIBs, irrespective of their monetary value. However, it was found in a study that Life Cycle Assessment (LCA)-aided Circular Economy (CE) approaches have not found much space yet in the literature. Exploring these studies can help in better methodological choices and give better results for research [158].

10. Conclusion

Spent LIBs rendered unattended in landfills are extremely harmful, leading to environmental degradation. Proper management of LIB wastes targeted with the circular economy is the only solution for spent lithium-ion batteries. This paper offers a comprehensive exploration of all aspects of spent LIB types, their environmental impacts, and recycling studies promoting a circular economy and research required in the area of recycling of spent LIB. The studies indicate that NMC batteries are the most commonly used battery type out of the six most used battery types (i.e., LFP, NMC, LCO, NCA, LMO, and LTO) primarily because of their high energy density and safety. The usage of LFPs has constantly declined due to their heaviness, low energy density, and low voltage. The suitability of recycling techniques depends upon the chemical makeup of the battery, which differs significantly. From the literature, it becomes evident that cathodes form a major part of such batteries, thus justifying the sustained focus on cathodes while recycling.

The literature review reveals that although the pyro metallurgical process is applicable at a large scale, it is energy-intensive, causes excessive emission of toxic gases, and is not efficient at recovering heavy metals like lithium. Hydrometallurgical processes do not take up as much energy as pyrotechnical methods do. However, it is also associated with the generation of huge amounts of waste streams that are highly corrosive in nature because of the use of various types of organic as well as inorganic acids used in leaching the black mass obtained from LIBs, questioning its industrial applicability.

Direct recycling seems to be the best choice in the available literature, as it requires the least amount of energy and barely generates hazardous secondary wastes. However, it demands additional treatment steps to obtain properly recoverable metals. Thus, achieving a circular economy for lithium-ion batteries requires a multi-faceted approach. This includes designing batteries with recyclability in mind, implementing efficient collection and sorting systems, developing innovative recycling technologies, and fostering collaboration among stakeholders. Prioritizing and implementing efficient recycling policies is essential for effective LIB waste management. For promoting a circular economy, the implementations demand the availability of a collection infrastructure for spent LIB and incentivizing recycling. More research focusing on sustainable recycling and material substitution is the need of the hour that would reduce the reliance on scarce mineral resources, ensuring sustainability. Supporting regulations are crucial for reintegrating recycled materials into new LIBs, fostering a circular economy. This comprehensive, policy-driven approach would minimize the environmental impact and guarantee sustainable energy storage. The implementation of circular economy principles ensures the minimization of waste and optimized resource utilization, creating a sustainable and resilient battery ecosystem. Considerable research is required to propose the most efficient and sustainable technique,

emphasising a holistic approach to LIB recycling within a circular economy framework.

The detailed literature review also reveals several operations challenges in spent LIB recycling. There is a lack of an all-component integrated approach intended to value all waste fractions; very few research have looked at the recovery of the other materials included in LFP batteries, with the recovery of Li still receiving the majority of attention. Also, the entire value chain is impacted by improper labelling since it is difficult to determine what kind of cathode enters the treatment facility, which reduces recycling effectiveness and product purity as most of the recovery methods are dependent on the chemical composition of the battery being handled. Recycling facilities must adapt to handle mixed-type cathodes and a wide variety of LIB scrap comprising diverse chemistries. Even while studies on recycling and valuation are becoming more prevalent, most of them are only evaluated in laboratories, and there are few commercial recycling methods. Therefore, more pilot work is required to scale up these technologies to an industrial level. These initiatives should be paired with sustainable battery production methods, the use of renewable energy sources while producing batteries, and the goal of extending the lifespan of battery systems. These, combined with advancements in battery technology, may result in a significant decrease in the cradle-to-grave LIB footprints.

Declaration of competing interests

The authors declare that they have no competing interests.

Authors contribution

Aparna Mukherjee: Data collection and manuscript writing, Sayali Apte: Conceptualization, supervision and review, Sayali Sandbhor – Supervision and review, Daniel Deng Kiir Kuany - Review

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