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Recovery Approaches for Spent Batteries: A Review

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Abstract
Lithium-ion batteries (LIBs) are being used in several applications such as consumer gadgets, electric vehicles, and renewable energy storage systems now days. This widespread use of these batteries has increased battery waste and created severe environmental and financial issues. Considering these issues, this review article is focus to throw light on the existing methods for recovering spent batteries. These methods are effective in recovering precious components including graphite, manganese, lithium, cobalt, and nickel making them available to reuse. Thus, these methods are able to resolve both environmental and financial crises. The review also discusses these recovery systems' scalability, economic feasibility, and safety factors, offering insights into the potential futures for battery recycling technologies. This review seeks to support the advancement of sustainable and effective recycling techniques for spent batteries by assessing the most recent recovery approaches and highlighting important opportunities and challenges. In the end, this will help to promote the circular economy and lessen the environmental impact of battery production and disposal.

Keywords- Spent batteries, Recovery techniques, Pre-treatment, Lithium ion battery.

1. Introduction

Lithium-ion batteries (LIBs) have a wide range of uses, including electronic devices, electric cars, and energy storage systems. The development of $LiCoO_2$ as the active cathode material was attempted and accomplished by Goodenough et al in 1979 (Mizushima et al., 1980). Later in 1991–1992, SONY and a joint venture between Asahi Kasei and Toshiba successfully commercialized the first LIB (Ozawa, 1994). The world's total lithium supply exceeded 634,000 metric tons in 2022. By 2030, it is anticipated that the global lithium supply would reach 2.14 million metric tons. There will still likely be a shortage of 2.3 to 2.45 million metric tons compared to the projected demand for lithium in 2030, even if the supply of lithium is predicted to increase by more than three times between 2030 and 2022 (Xing and Srinivasan, 2024). For production to maintain up with the anticipated yearly rises in worldwide requirements for lithium over the decade that follows, significant lithium deposits must be brought online each year. In 2023, India may produce 18 gigawatt hours of LIBs. The value is expected to increase to more than 150 gigawatt hours by 2030 (Deorah et al., 2020).

With increase in the usage of rechargeable batteries, there is lot of concern for the utilization of spent batteries (Jena et al., 2021). Concerns over resource depletion and the effects on the environment have been highlighted by the rising demand for LIBs. Recovering valuable resources, lowering environmental risks, and promoting the circular economy all depend on recycling wasted LIBs. For the preservation of the environment and sustainable resource management, spent lithium-ion battery recycling is essential. The recycling of spent batteries has gained attention among research community because of the raising alarm of the depletion and shortage of raw materials of battery components. As illustrated in **Figure 1 (a)**, research trends on spent batteries have rapidly increased between 2010 and the present and the inset shows the type of articles published. The keywords used for collecting the data are represented in the **Table 1**. Considering the concern over increased solid waste with the development of rechargeable batteries and countries are making efforts in this direction taking China as lead (Mrozik et al., 2021; Porzio and Scown, 2021; Noudeng et al., 2022; Tang et al., 2023). Though USA and India are just after China but their efforts in these directions are less compared to China as shown in **Figure 1 (b).** Considering the case of India, it is producing almost 1/9 times to China, which shows the necessities of research in this direction (Kala and Mishra, 2021; Deshwal et al., 2022). Work in this direction is not only limited to reducing solid waste but also helpful to make the devices more economically sounds. **Figure 1 (c)** shows the global distribution of common LIB recycling enterprises.

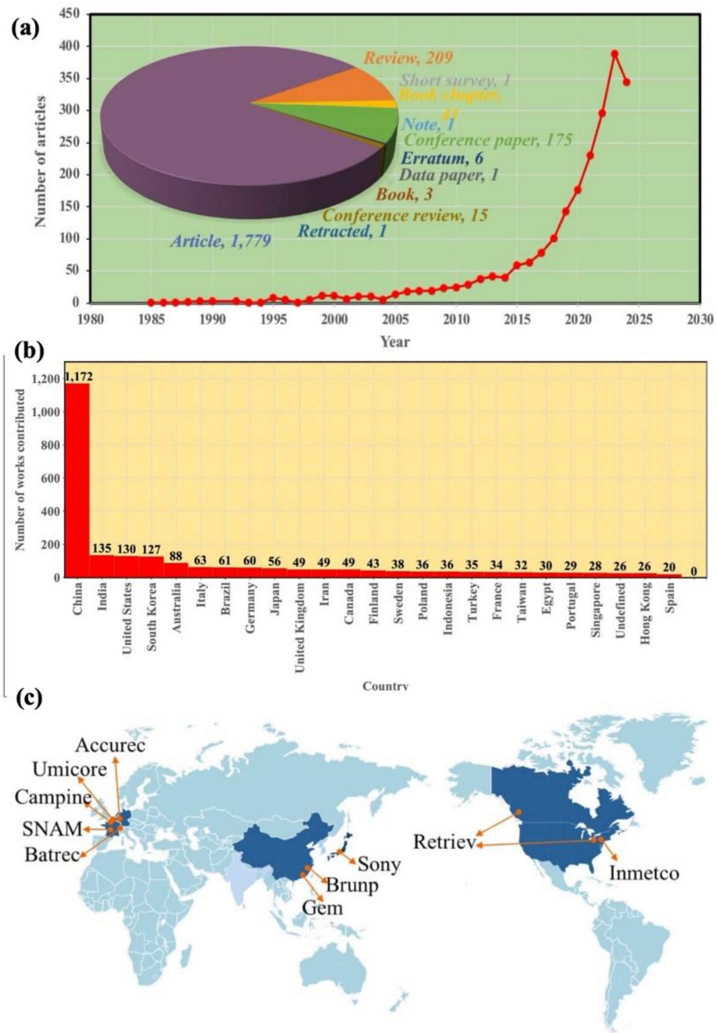

Figure 1. (a) Number of articles published from 1985 to August 2024 related to recovery of spent battery and inset shows the type of articles published, (b) Contribution of different countries. Data is taken from the Scopus as on August 2024, (c) Global distribution of common LIB recycling organizations (Li et al., 2018).

It is important for resource conservation and environmental sustainability that valuable materials, especially lithium-ion batteries (LIBs), be recovered from wasted batteries. Recent studies have concentrated on several approaches for recovering important metals such as nickel, cobalt, manganese, and lithium from old batteries. For example, Cheng et al. (2024) emphasizes improvements in the hydrometallurgical and pyro-hydrometallurgical processes for selectively extracting lithium from used LIBs. Similar to this, Li et al. (2023) provides a thorough analysis of recycling technologies that takes into account both the effectiveness of resource recovery and the effects on the environment. Guo et al. (2024) concentrated on LIBs that contain manganese and talk about different recovery strategies such hydrometallurgical and bioleaching procedures that strike a compromise between environmental sustainability and economic viability. Furthermore, the recovery of electrolytes and cathode materials is examined by Zheng et al. (2024). Authors have compare the environmental effect and efficiency of pyrometallurgical and hydrometallurgical processes, indicating that hydrometallurgy might provide more environmentally friendly options. Jing et al. (2024) investigated the reuse of lithium and iron from lithium iron phosphate batteries by emphasizing on the possibility of direct regeneration techniques and mechanochemical processes for preserving material integrity and minimizing environmental impact. Together, these studies highlight how crucial it is to create scalable, environmentally benign, and effective solutions for recovering wasted batteries in order to support a long-term circular economy within the battery sector. Though different kind of batteries exist now a days but this work is focussed on Li rechargeable batteries based on nickel, manganese, cobalt oxide cathode (Singh, 2023; Singh et al., 2024). The procedures and technologies used to recycle these batteries are outlined in this review.

Table 1. List of keywords for data collections from Scopus as of August 2024.

Results	Keyword	Results	Keyword
1,240	Lithium-ion Batteries	81	Kinetics
1,172	Article	80	Hydrometallurgical Process
891	Cathodes	79	Spent LIBs
869	Cobalt	79	Activation Energy
816	Electronic Waste	78	Pyrolysis
763	Ions	77	Transition Metals
689	Leaching	77	Deep Eutectic Solvents
649	Lithium	75	Unclassified Drug
635	Lithium Compounds	75	Selective Recovery
541	Metal Recovery	74	Thermodynamics
460	Recovery	73	Regeneration
443	Recycling	73	Rare Earths
372	Spent Lithium-ion Batteries	73	Lead Acid Batteries
301	Manganese	73	Hydrogen Peroxide
299	Cathodes Material	73	Bioleaching
285	Cobalt Compounds	73	Ammonia
285	Electric Batteries	72	Lithium Recoveries
259	Extraction	70	Eutectics
245	Hydrometallurgy	69	Cadmium
239	Lithium Ion	68	Ion
215	Metals	68	Heavy Metals
211	Nickel	67	Leaching Solution
205	Electrodes	67	Environmental Protection
201	Anodes	66	Slags
192	Electric Power Supplies	65	Citric Acid
191	Iron Compounds	64	Electrochemical Performance
188	Manganese Compounds	64	Chemistry
186	Nickel Compounds	63	Waste Treatment
176	Power Supply	63	Leaching Process
172	Scanning Electron Microscopy	63	Environmental Pollutions
171	Solvent Extraction	63	Crystal Structure
168	Spent Batteries	62	Carbon Dioxide

Table 1 continued…

165	Temperature	62	Adsorption
162	Valuable Metals	61	Electric Battery
158	Calcination	60	Reduction
156	Precipitation (chemical)	59	Scrap Metal Reprocessing
155	Zinc	59	Life Cycle
155	Graphite	59	Heat Treatment
152	Battery Recycling	58	Nickel Metal Hydride Batteries
145	Controlled Study	58	Electronic Equipment
143	X Ray Diffraction	57	Sodium Compounds
143	Sulfur Compounds	57	Oxygen
142	Metal Ions	56	Sodium Carbonate
140	Sulfuric Acid	56	Selective Extraction
139	Separation	56	Recycling Technology
138	Manganese Oxide	81	Lithium Alloys
138	Efficiency	56	Procedures
126	Sustainable Development	56	Extraction Method
126	Electrode	56	Cathode Active Material
126	Chlorine Compounds	55	Metals Recoveries
118	Metal	55	Lead
118	Energy Utilization	55	Flotation
117	Carbon	55	Environmental Technology
114	Recovery Of Valuable Metals	55	Electric Discharges
113	Precipitation	55	Cost Effectiveness
113	Electrolytes	53	Organic Acids
110	Lithium-ion Battery	53	Nickel Oxide
107	Aluminium	52	Secondary Batteries
105	Iron	51	Water Leaching
104	Metallurgy	51	Waste Disposal
102	Recycling Process	51	Selective Leaching
98	Copper	51	Fourier Transform Infrared Spectroscopy
97	Spent Lithium-ion Battery	51	Circular Economy
97	Solvents	51	Chemical Analysis
97	Priority Journal	50	Spent Lead Acid Batteries
96	Lithium Batteries	50	Nonhuman
95	Environmental Impact	49	Recovery Efficiency
95	Dissolution	49	Ion Exchange
93	Particle Size	49	Anode Material
93	Cathode Materials	48	Purification
92	Oxalic Acid	48	Lithium Ions
91	Pyrometallurgy	48	Electrolysis
89	Waste Management	48	Chemical Reaction
86	Oxidation	48	Alkalinity
85	Energy Storage	47	Lithium Recovery
85	Electrode Material	47	Electrochemistry
83	Acid Leaching	47	Chemical Composition
82	Reaction Kinetics	46	Carbothermal Reduction
81	Sodium Hydroxide	46	Battery Industry
81	PH	46	Alkaline Batteries

2. Components of a Battery

In order to store and release electrical energy, a battery is made up of various essential principal elements like anode, cathode, separator, electrolyte, current collectors and casing. **Figure 2** shows the components and their corresponding concentrations in LIB.

- ***Anode*** **(*Negative Electrode*):** During the discharge process, the anode of the battery releases electrons and materials like lithium, graphite, or zinc are used to make it.

- ***Cathode* (*Positive electrode*):** During the discharge process, the cathode is the part that receives electrons and materials like manganese dioxide, nickel, or lithium cobalt oxide are frequently used to make it.

- ***Electrolyte*:** An electrolyte is a medium that facilitates the flow of electrical current by allowing ions to migrate between the anode and cathode. Depending on the kind of battery, the substance may be liquid, gel, or solid.

- ***Separator*:** To prevent the anode and cathode from coming into direct contact, which could result in a short circuit, the separator is a porous membrane that physically divides them. It obstructs the flow of electrons within the battery but permits the movement of ions.

- ***Current Collectors*:** Electrons are collected and transferred to an external circuit by means of these conductive materials, which are coupled to the anode and cathode. Aluminium is typically used for the cathode current collector and copper for the anode current collector.

- ***Casing*:** The battery's exterior shell, or casing, encloses every component to keep it safe from harm and to contain any internal chemical reactions.

The cathode material is the standard method of classification for LIBs (Sobianowska-Turek et al., 2021). The choice of cathode is crucial based on the particular application since different cathode types offer varying balances of energy density, power output, longevity, cost, and safety. Some of the cathode materials widely being used in LIBs are $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, $LiMn_2O_4$, $LiNiMnCoO_2$, $LiCoO_2$, $LiFePO_4$ (Fu et al., 2023). **Table 2** shows the elemental and material mass concentrations of various cathode materials in LIB based on the data derived from the reported literature (Winslow et al., 2018).

Table 2. Elemental and material concentration in percentage for (a) $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ cathode (b) $LiMn_2O_4$ cathode (c) $LiNiMnCoO_2$ cathode (d) $LiCoO_2$ cathode (e) $LiFePO_4$ cathode of LIB samples. Values are adopted from Winslow et al. (2018).

Element/Materials	Concentration in percentage				
	(a)	(b)	(c)	(d)	(e)
Oxygen	8	12	5	-	-
Plastics	4	5	3	5	4
Aluminium	22	22	23	5	7
Cobalt	2	-	8	17	-
Copper	13	14	17	7	8
Lithium	2	1	1	2	1
Manganese	-	11	6	-	
Nickel	12	-	15	1	
Binder	4	4	1	2	1
Iron/Steel	0	0	9	-	43
Carbon (non-graphite)	2	2	3	6	2
Electrolyte + Solvent	12	12	2	14	15
Graphite	17	16	-	23	13
Thermal Insulation	1	1	-	-	
Fluoride	-	-	5	-	
Phosphorus	-	-	2	-	5

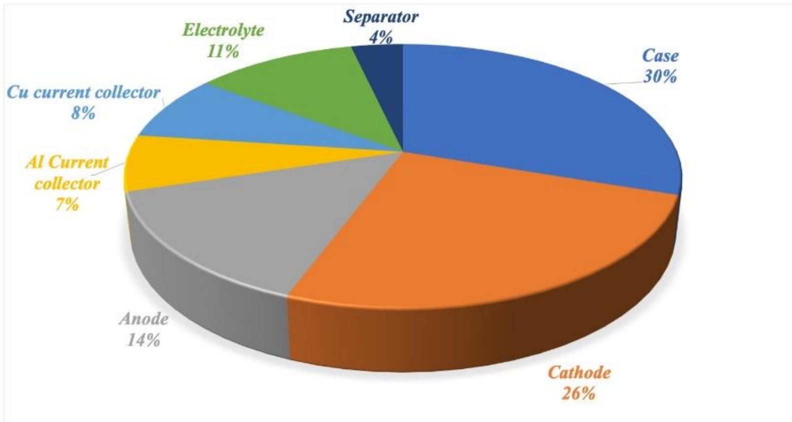

Figure 2. Components of a battery such as anode, cathode, separator, electrolyte, current collectors, and casing.

3. Recycling Methods for Spent Li-ion Batteries

Recycling LIB cathode materials is a crucial procedure for a number of reasons, including cutting production costs, protecting the environment, and saving valuable resources. Resource recovery is much important as the valuable and scarce metals like lithium, cobalt, nickel, etc are present in LIBs. Recycling aids in the recovery of these metals, hence lowering the necessity for mining and the negative effects it has on the environment and society (Khorami et al., 2019). Recycling lessens the quantity of hazardous garbage that would otherwise find its way into landfills, where it might release harmful substances into the atmosphere. Additionally, it lessens the carbon impact brought on by the extraction and refinement of new materials. Advancement in battery technology is fuelled by the need for more economical and efficient recycling techniques, which results in the creation of batteries that are simpler to recycle or that make use of more widely available, safer materials. Manufacturers can maintain high standards in new batteries, possibly increasing performance and prolonging battery life, by reusing and refining cathode materials. **Figure 3** shows the various recycling methods available for spent LIBs.

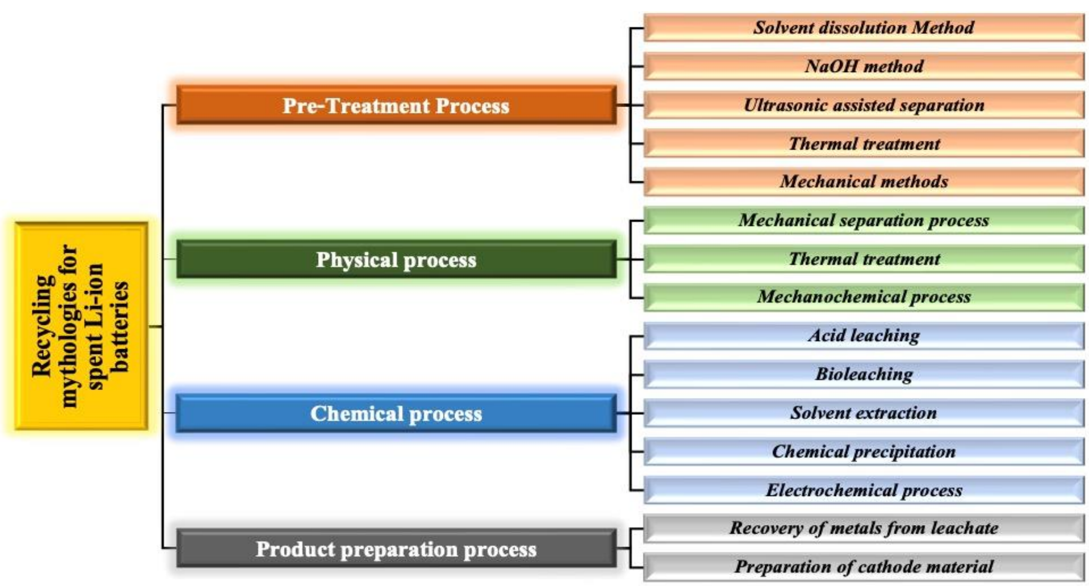

Figure 3. Recycling methods available for spent lithium-ion batteries.

3.1 Pre-Treatment Process

LIB recycling usually consists of the steps like material recovery, disassembly, collection, discharge, and separation **Figure 4**. Batteries are first gathered from end users via a variety of channels, including dealerships, electronic waste collection places, and targeted recycling programs. As LIBs can entail risks including heat runaway and chemical leakage, safe transporting procedures are crucial (Qiu and Jiang, 2022). In order to reduce the possibility of electrical shock and fire during handling, spent batteries are completely drained (Torabian et al., 2022). Batteries can be disassembled manually or mechanically to separate parts such the separator, electrode, electrolyte, and casings. Later they are subjected to various recycling processes like physical process, chemical process and product preparation process.

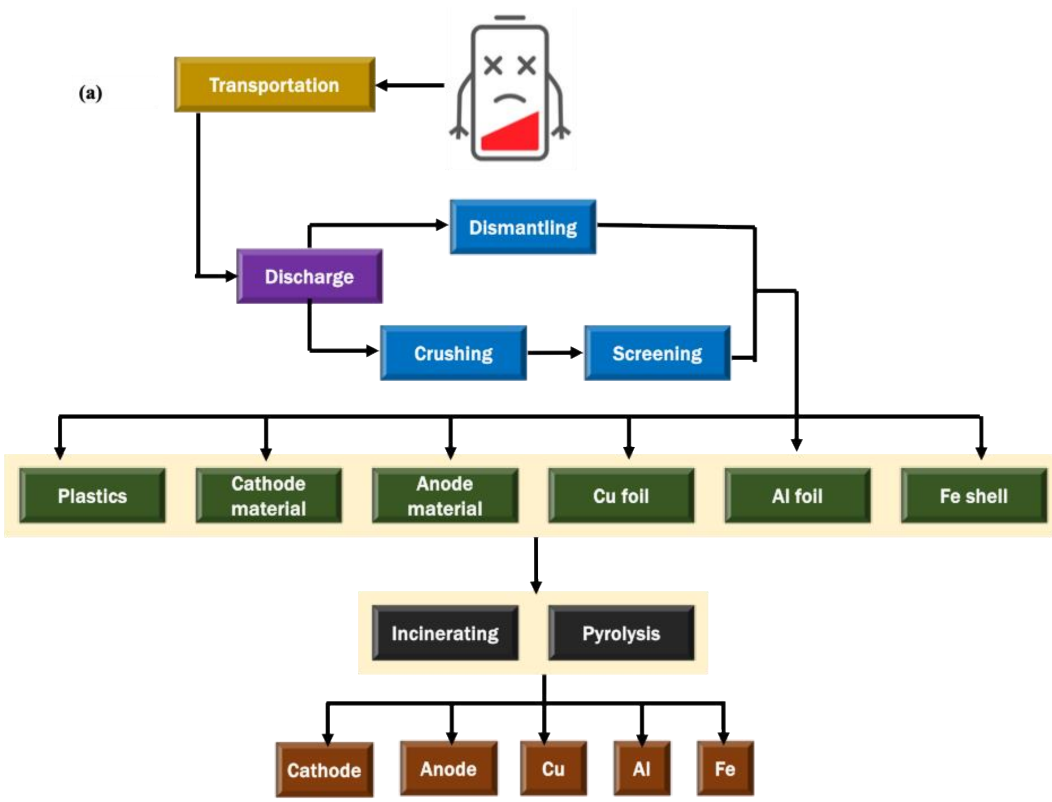

Figure 4. Pre-treatment process.

3.1.1 Solvent Dissolution Method

When it comes to LIBs, which are extensively utilized in electronics, electric cars, and energy storage systems, the solvent dissolution method (**Figure 5**) is crucial to the recovery and recycling of valuable elements from wasted batteries. The principal objective is to reduce waste and environmental impact while carefully dissolving and recovering valuable metals and active elements, including as lithium, cobalt, nickel, manganese, and graphite, from the battery components. The cathode materials are subjected to an appropriate solvent as the process weakens the adhesion of the PVDF which holds the electrode active materials to the current collectors (Ordoñez et al., 2016). For example, $LiCoO_2$ or other cathode materials can be dissolved using an acidic or organic solvent. Metal oxides in battery cathodes are frequently dissolved using strong acids, such as sulfuric acid (H_2SO_4) or hydrochloric acid (HCl), and organic solvents, such as dimethyl sulfoxide (DMSO) or ethylene carbonate (Junior et al., 2024). Subsequent research on dissolving process is focussed towards the employment of solvents that are more eco-friendly, like supercritical CO_2 or ionic liquids (Shi et al., 2024).

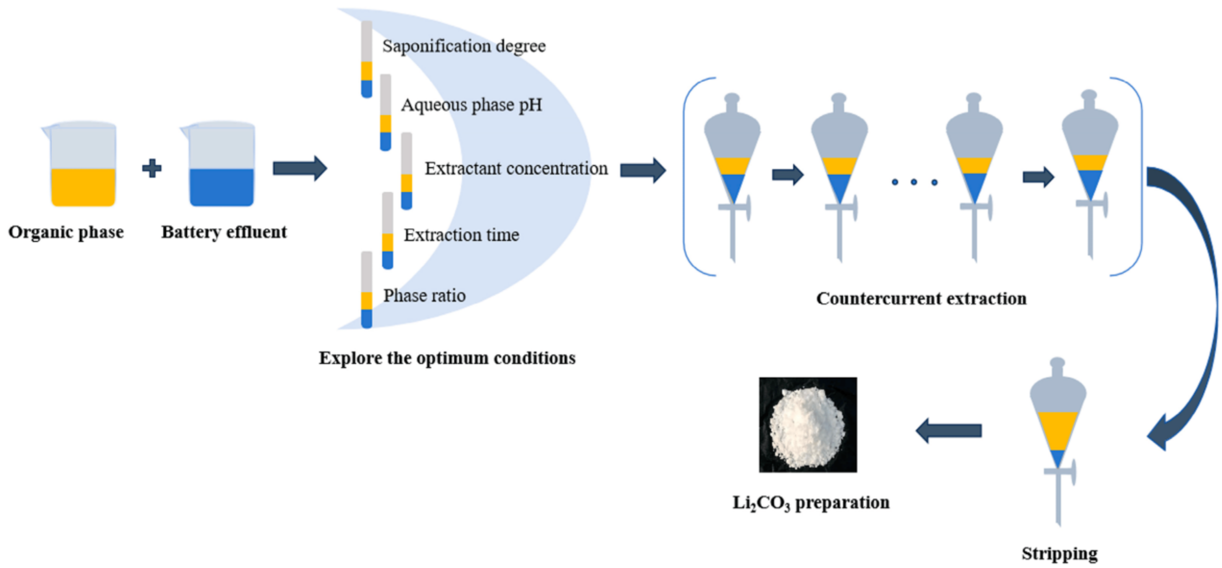

Figure 5. Schematics for the solvent dissolution method (Wang et al., 2024; open access).

3.1.2 NaOH Method

The NaOH method is used to extract or purify specific elements from the spent battery components. This technique utilizes aqueous chemistry and is frequently a component of a larger hydrometallurgical process. Following the mechanical separation of parts of batteries in LIB recycling, the cathode material is frequently treated using NaOH for dissolving the aluminium foil. Aluminium foil is dissolved into sodium aluminate ($NaAlO_3$) by reaction with NaOH, while the active cathode material remains intact. The other important metals like cobalt, nickel, and lithium can be extracted from the residual active cathode material using techniques like acid leaching. In many battery-recycling procedures, the NaOH technique is an essential step that helps with the effective and selective recovery of essential elements (Cai et al., 2014) and detailed recovery procedure for spent $LiFePO_4$ is elaborated by Li et al. (2022). This technique promotes a sustainable supply of vital raw materials and lessens the negative environmental effects of battery waste. Because it is widely available and cost-effective, sodium hydroxide is an economical reagent for large-scale battery recycling operations and it is very good at dissolving aluminium in a selective manner.

3.1.3 Ultrasonic Assisted Separation

With the demand for battery materials advancing, it is imperative to increase the efficacy and efficiency of the recycling process and ultrasonic assisted separation is a vital tool in achieving this goal. By using high-frequency sound waves (usually between 20 kHz and several MHz), ultrasonication can produce cavitation bubbles in a liquid media (Paniwnyk, 2014). Rapid formation and collapse of these bubbles results in extremely high local temperatures and pressures. This phenomenon generates shock waves and microjets that can accelerate mass transfer rates, improve chemical processes, and mechanically disturb materials. By inducing vibrations that lessen the adhesive forces holding both the electrode content and foils together, the ultrasonic waves successfully separate them. Either an aqueous or solvent-based media can be used for this procedure. Ultrasonication can greatly improve leaching efficiency, which is the process by which metals are dissolved from the materials used in electrodes using acids or other solvents (Shih et al., 2019; Zhao et al., 2020). Solid particles are broken down by the cavitation action, which increases the amount of surface area available to the leaching agent. Higher rates of valuable material recovery can result from the separation and extraction processes accomplished much more quickly through ultrasonication (He et al., 2015). Although ultrasonication works well in lab environments, it can be difficult to scale up for use in industrial settings, particularly when it comes to distributing energy uniformly throughout vast volumes of material.

3.1.4 Thermal Treatment

Thermal treatment technique adds to the general environmental sustainability of battery recycling processes by carefully managing the heat used to old batteries in order to remove dangerous components and condition materials for effective recovery (Pigłowska et al., 2024). Organic binders, like as polyvinylidene fluoride (PVDF), hold together a lot of battery components, particularly the electrodes. These binders can be broken down by heat pre-treatment. This facilitates the removal of the active ingredients from current collectors. In this process, LIBs are typically disassembled and then heated to approximately 300°C as part of the pre-treatment step (Tawonezvi et al., 2023). This temperature is high enough to evaporate the electrolyte and break down organic binders without melting the metal components. Following cooling and mechanical separation of the treated materials, the active materials are gathered for additional processing. **Figure 6** shows the schematic diagram of the recovery process for spent batteries by thermal treatment.

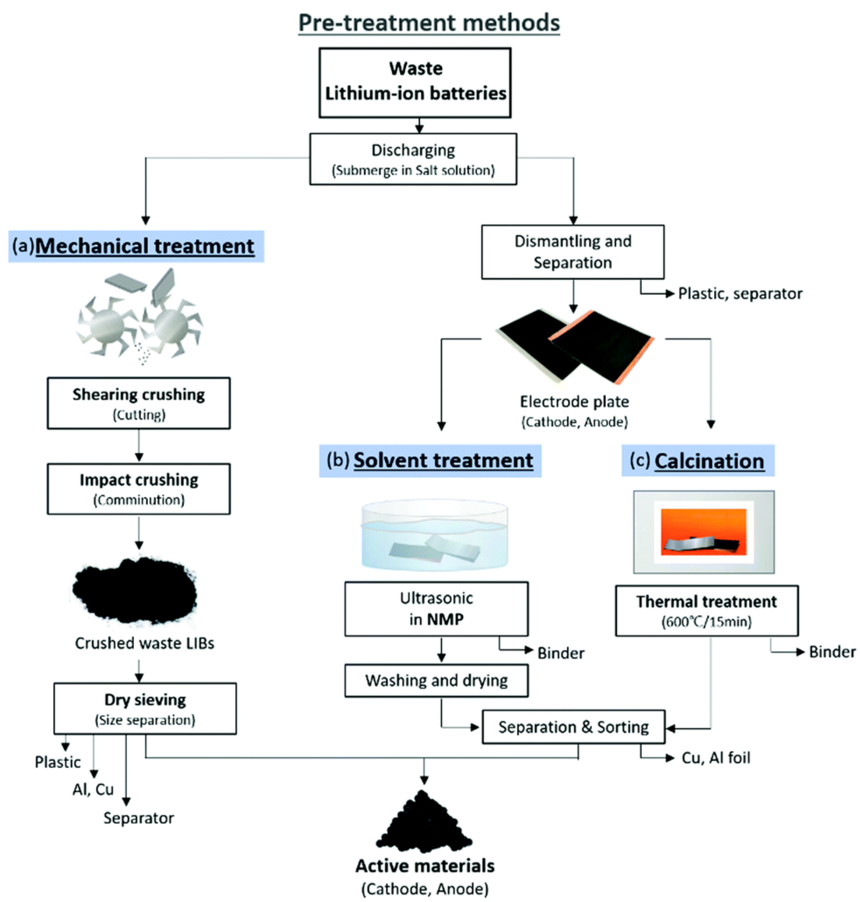

Figure 6. Schematic diagram of the recovery process for spent batteries by mechanical, solvent and thermal treatment (Bae and Kim, 2021; open access).

3.1.5 Mechanical Methods

A key phase in the recovery of battery material is mechanical pre-treatment, which makes it possible to effectively separate and concentrate valuable components. Mechanical pre-treatment categorizes batteries into their component pieces and efficiently sorts these materials, paving the way for the effective recovery of metals along with additional elements in later recycling steps. Batteries are broken down into tiny pieces by first being crushed or shredded. Materials like polymers, current collectors, active electrode materials, and other components are liberated during this process. Battery components are made diminished by crushing, which also helps to reveal the chemical components for simpler separation in later stages. The material mixture is usually run through screens or sieves to separate the particles according to size after shredding or crushing. Larger metal fragments are helped to be separated from smaller particles containing active components by this phase so that they can be treated further. Steel casings and other ferromagnetic materials are separated from the broken battery components using magnetic separation. Metal components can be effectively separated from non-magnetic materials like plastics and active powders using this method (Smith et al., 2019). Complete material separation can be difficult to accomplish due to the physical properties of battery components, resulting in mixed fractions that can need additional processing. combining thermal, chemical, and mechanical pre-treatment techniques into a single, efficient process to increase material recovery and decrease waste. **Table 3** shows the overall classification of pre-treatment techniques along with their advantages and challenges.

Table 3. Classification of pre-treatment techniques along with their advantages, challenges, applications and emerging trends.

Method	Advantages	Challenges
Solvent Dissolution Method	• High Efficiency • Selectivity • Compatibility	• Solvent recovery • Handling of Hazardous solvent • Scalability
NaOH Method	• Selective dissolution • Cost effective • Environment friendly	• Waste Management • Material Specificity
Ultrasonic Assisted method	• Increased Efficiency • Selective Separation • Environment friendly	• Energy consumption • Scaling up • Equipment costs
Thermal Pre-Treatment	• High Recovery Efficiency • Volume Reduction • Hazardous Material Neutralization	• Energy intensive • Environmental concerns • Loss of valuable material
Mechanical Pre-Treatment	• Safety • Improved Recovery Efficiency • Volume Reduction	• Energy consumption • Loss of valuable material • Emission control

3.1.6 Pyrometallurgy

One of the main techniques for recycling used in LIBs is pyrometallurgy which uses high temperatures to remove valuable metals from battery trash, including iron, nickel, copper, and cobalt. Because pyrometallurgy can tolerate impurities and manage varied battery chemistries, it is commonly employed. Usually, the procedure is done in phases, such as pre-treatment, smelting, and refining (**Figure 7**). The prepared battery substances are put into a high-temperature furnace during the smelting stage, which usually operates between 1,200°C and 1,600°C (Qu et al., 2023). Depending on the intended results, the furnace atmosphere can be either oxidative or reductive. Metals such as copper, nickel, and cobalt undergo reduction after oxidation to create an alloy rich in metals, while aluminum, lithium, and other lighter elements produce a slag in an oxidative atmosphere. Reducing agents such as carbon or coke are applied in a reductive environment to help reduce metal oxides to their metallic states. Metal-rich fractions can be easily separated during smelting because of the heavier metal alloy's separation from the lighter slag caused by differences in density. The metal alloy is refined to extract individual elements and further purify the metals after smelting. During this phase, hydrometallurgical methods like solvent extraction, electrolysis, or leaching may be used to extract and purify certain metals like copper, nickel, and cobalt. Following procedures like leaching or roasting can extract lithium, which usually ends up in the slag. In order to produce high-purity metals that can be used again in the manufacturing of new batteries or other applications, the refining stage is essential.

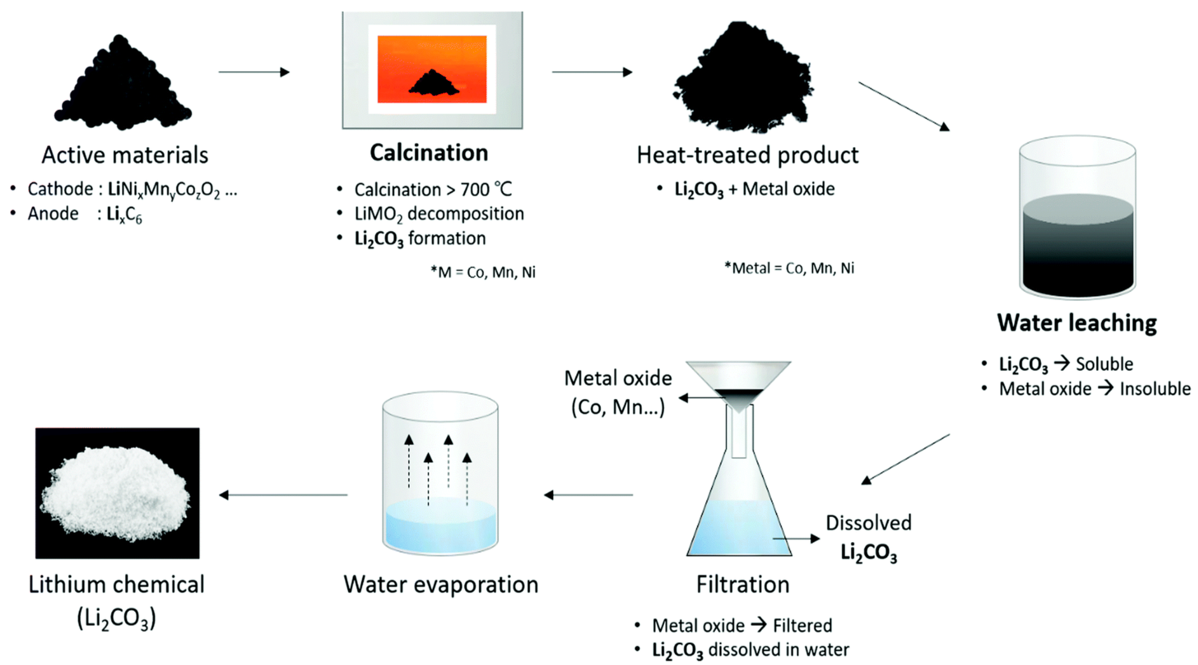

Figure 7. Schematic of recycling of LIB components by pyrometallurgy process (Bae and Kim, 2021).

In this technique, organic components like binders and electrolytes are effectively destroyed by the high temperatures utilized in pyrometallurgy. This lowers the possibility of harmful emissions. Additionally, the procedure is comparatively quick and may recover metals like copper, nickel, and cobalt at high rates. The pyrometallurgical method has a number of drawbacks despite its advantages. Smelting is an expensive and environmentally harmful process that contributes to greenhouse gas emissions due to its high energy consumption. Apart from this, the process produces slag, which poses environmental problems because to its residual metal content and need for additional processing or disposal (Potysz et al., 2018). Furthermore, aluminum and lithium are harder to recover using the pyrometallurgical method; instead, they usually end up in the slag and need extra processing.

Considering, it's drawback, a lot of attention is being provided to create more energy-efficient smelting technologies. Use of microwave or plasma smelting, followed by refining slag treatment procedures to maximize lithium recovery can be helpful. Furthermore, pyrometallurgy and hydrometallurgical processes can be integrated to minimize environmental effects and maximize overall metal recovery rates.

3.1.7 Mechanochemical Process

The mechanochemical process, in contrast to conventional physical or chemical separation techniques, uses mechanical force to directly cause chemical reactions in the solid state, frequently at room temperature and without the need for additional heating or solvents. Because of its potential for lower energy usage, a less environmental effect, and higher metal extraction efficiency, this makes it an attractive technology for LIB recycling. The mechanical energy used in high-energy ball milling or comparable processes is what drives the mechanochemical process, which breaks down the crystal structures of the electrode materials. Intense mechanical forces during this procedure cause flaws and increase the materials' surface area, which promotes chemical reactions and the liberation of precious metals. The direct synthesis or breakdown of materials is facilitated by the mechanical action's ability to overcome the activation energy barriers of chemical reactions, which eliminates the need for harsh chemicals or high temperatures. The mechanical energy of the materials being ground promotes a variety of chemical reactions. For instance, active metals can be released from their oxide matrices by mechanochemically treating wasted LIB cathodes, such as lithium cobalt oxide ($LiCoO_2$) or lithium nickel manganese cobalt oxide (NMC). Depending on the conditions, this process can directly produce metal salts, oxides, or other compounds. During milling, additives like carbon or other reducing agents may be added to increase reaction rates or encourage particular reactions (Wang et al., 2021).

Comparing the mechanochemical approach to traditional recycling techniques reveals a number of benefits. By doing away with the need for harsh chemicals and high temperatures, it lowers energy usage and its impact on the environment. By modifying the milling settings and chemical additives, the technique can also selectively extract particular elements and obtain excellent recovery rates for essential metals. It is especially appealing for recycling a variety of LIB formulations and designs because of its flexibility.

To reach its full potential in commercial applications, the mechanochemical process must overcome several obstacles in addition to its benefits (Dolotko et al., 2023). The process can be scaled up to handle huge volumes of wasted batteries, the efficiency of the leaching and metal recovery procedures that follow can be increased, and the milling parameters can be optimized to maximize the recovery of metal.

3.2 Chemical Process

3.2.1 Acid Leaching

Acid leaching is a frequently used procedure in the recycling of used LIBs to recover important metals like lithium, cobalt, nickel, and manganese. The metal elements of the battery's working elements, mainly the cathode, are dissolved using acidic liquids, usually sulfuric acid. The battery material is subjected to regulated acid treatment during the leaching process, which dissolves the metals and forms metal salts in the process. Subsequent processing of this leachate can precipitate the metals, which are then refined and put to use in the creation of new batteries or other uses. The significance of acid leaching lies in its capacity to successfully extract vital metals from scrap batteries, which are normally hard to separate and recover through mechanical means alone. It additionally serves a key part in the sustainable utilization of battery materials by reducing the need for virgin metal mining, saving natural resources and minimizing environmental impact.

The use of organic acids, deep eutectic solvents, and synergistic leaching techniques are some of the innovations in the field of sustainable metal recovery from used batteries (Wang et al., 2022). Sulfuric acid and hydrogen peroxide are common chemicals in traditional leaching procedures, which are criticized for having a negative environmental impact despite producing high recovery rates for metals including cobalt, nickel, and lithium. Innovative strategies aim to figure out these issues. For example, synergistic leaching mixing sulfuric and organic acids, such as citric acid, has been found to successfully remove metals without the requirement for reductants, improving both efficiency and environmental safety (Roshanfar et al., 2024). Additionally, the use of deep eutectic solvents, particularly those based on pyruvic acid, has exhibited extraordinary effectiveness, extracting over 99% of Co and Li while decreasing hazardous by products (Islam et al., 2021; Chenthamara and Gardas, 2024). Additionally, glutamate leaching offers a novel strategy that emphasizes the significance of pH in maximizing recovery rates and decreasing acid consumption, especially in neutral settings (Prasetyo et al., 2024). These developments represent a substantial leap in battery recycling technology since they not only enhance metal recovery but also lessen their negative effects on the environment.

3.2.2 Bioleaching

Bioleaching uses the natural metabolic activities of some bacteria and fungi to solubilize metals from the battery components, as opposed to standard acid leaching (Desmarais et al., 2020), which uses powerful acids and harsh chemicals (Roy et al., 2021). In this method, spent battery material is mixed with a microbial culture in a bioreactor or leaching tank under carefully controlled conditions, such as temperature, pH, and oxygen levels (Mishra and Rhee, 2014). Microorganisms like *Acidithiobacillus ferrooxidans*, *Leptospirillum ferrooxidans*, and *Acidithiobacillus thiooxidans*, that are capable of converting metal sulfides into soluble metal sulfates, can be employed to break down the metal compounds in battery cathodes and release metals like lithium, cobalt, nickel, and manganese into solution (Annamalai and Gurumurthy, 2019; Bajestani et al., 2014; Singh et al., 2018). When compared to traditional acid leaching, bioleaching has less of an impact on the environment. It produces fewer hazardous by-products by minimizing the use of powerful acids and chemicals, which facilitates the management and treatment of the waste. Time efficiency is crucial in industrial-scale processes, and bioleaching can be limited as it often takes more time than chemical leaching (Alipanah et al., 2023; Petersen, 2023).

Because different microbial agents have different metabolic processes and produce different types of acids, there are considerable variances in the efficacy of bioleaching in retrieving metals from used LIBs. Processes involving LIBs for remarkable-purity lithium carbonate recovery might benefit greatly from the high leaching efficiency of bacterial agents such as *Acidithiobacillus ferrooxidans*, which can achieve dissolution rates of up to 99.9% when the right circumstances are met (Alhaqie et al., 2023). Redoxolysis, a process that these bacteria rely on to produce sulfuric acid, is essential in dissolving metals in the highly acidic conditions needed for efficient leaching. Conversely, organic acids like citric, oxalic, and gluconic acids are produced by fungal agents like *Aspergillus niger*. These acids are also effective, but they typically have less metal rates of recovery for substances like nickel and cobalt, with highest rates of 13% for nickel and 25.8% for cobalt; however, they excel in lithium recovery, with rates reaching up to 87.9% (Gerold et al., 2024). In addition to its versatility, fungal bioleaching offers advantages like resistance to hazardous components and a broad pH range of operation. However, because of the intricate biochemical interactions involved, fungal bioleaching is often slower (Biswal and Balasubramanian, 2023).

3.2.3 Chemical Precipitation

A crucial technique in battery recycling is chemical precipitation (Dhiman and Gupta, 2019). This technique extracts valuable metals from leach solutions made from dead batteries. In this process, insoluble metal complexes are created by adding precipitating agent to the aqueous solution containing dissolved metal ions as these are simple to filter out. Chemical precipitation is a particularly helpful method for recovering metals including lithium, cobalt, nickel, and manganese in the context of recycling lithium-ion batteries. To precipitate only the target metal and leave the others in solution, certain parameters like pH, temperature, and concentration must be satisfied as well as a particular precipitating agent used. By altering the pH and employing reagents such as hydrogen sulfide or sodium hydroxide, cobalt and nickel can be retrieved as their hydroxides or sulfides, respectively.

The efficiency of chemical precipitation for recovering metals from battery recycling procedures depends critically on pH control (Lv et al., 2018). The solubility of metal ions and the related precipitates is directly influenced by the pH of the solution, allowing for the selective recovery of particular metals. Due to their individual solubility products, different metal ions start to precipitate at different pH values when a solution's pH is changed (Ksp) (Haas et al., 2019). Thus, by carefully regulating the pH, metals such as cobalt, nickel, manganese, and lithium can be precipitated selectively during the recycling of lithium-ion batteries. While lithium stays soluble, metals like cobalt and nickel can precipitate as their hydroxides at lower pH values. For instance, nickel hydroxide precipitates at a slightly higher pH of approximately 9–10 compared to cobalt hydroxide, which precipitates at a pH of about 8–9 (Harvey et al., 2011). Manganese can precipitate as manganese dioxide at pH values between 10 and 11 if the pH rises even further. Due to its much higher solubility, lithium usually needs a pH of 12 or higher to start precipitating, most frequently as lithium carbonate in the presence of carbonate ions (Biswal et al., 2018). The sequential recovery of metals with high purity is made possible by this stepwise precipitation based on pH, which involves gradually adjusting the pH and adding the necessary reagents. Additionally, careful pH control limits the co-precipitation of undesirable metals, boosting the efficiency and cost-effectiveness of the recycling process.

3.2.4 Electrochemical Process

Electrochemical recycling process for spent LIBs has got attention in recent years because of its promise for high selectivity, efficiency, and environmental sustainability. Electro winning, electro dialysis, and electrochemical extraction are the main techniques that are used in this process (Du et al., 2023). This process enables the recovery of important metals, including nickel, manganese, cobalt, and lithium. If compared with conventional pyrometallurgical and hydrometallurgical processes, the electrochemical recycling has a number of benefits, including lower energy consumption, less chemical waste, and the capacity to recover metals at high purity levels (Zheng et al., 2018). This is beneficial because the growing manufacturing of electric cars and renewable energy storage devices is driving up demand for these metals.

Electrowinning entails depositing metals from the leached solution onto electrodes using an electric current (Liu et al., 2024). Cobalt and nickel can be deposited onto the cathode selectively by varying the applied voltage during the process. Lithium ions are extracted and recovered from the solution by electrodialysis, which makes use of ion-selective membranes and an electric field. A greater purity level can be attained by further refining these recovered metals using electrochemical extraction, which is necessary for the metals to be reused in the creation of new batteries. **Figure 8** shows the experimental setup and flow chart for the recovery of Co, Li, and graphite from spent $LiCoO_2$ batteries.

The application of innovative electrolytes and electrode materials to improve the electrochemical recycling process' efficiency has also been the subject of recent research. Examples of attractive research areas include the creation of environmentally friendly electrolytes that minimize secondary waste and the improvement of electrode materials for increased conductivity and selectivity. Scaling up these procedures for industrial use is still difficult, especially when handling the various chemistries and designs of discharged batteries.

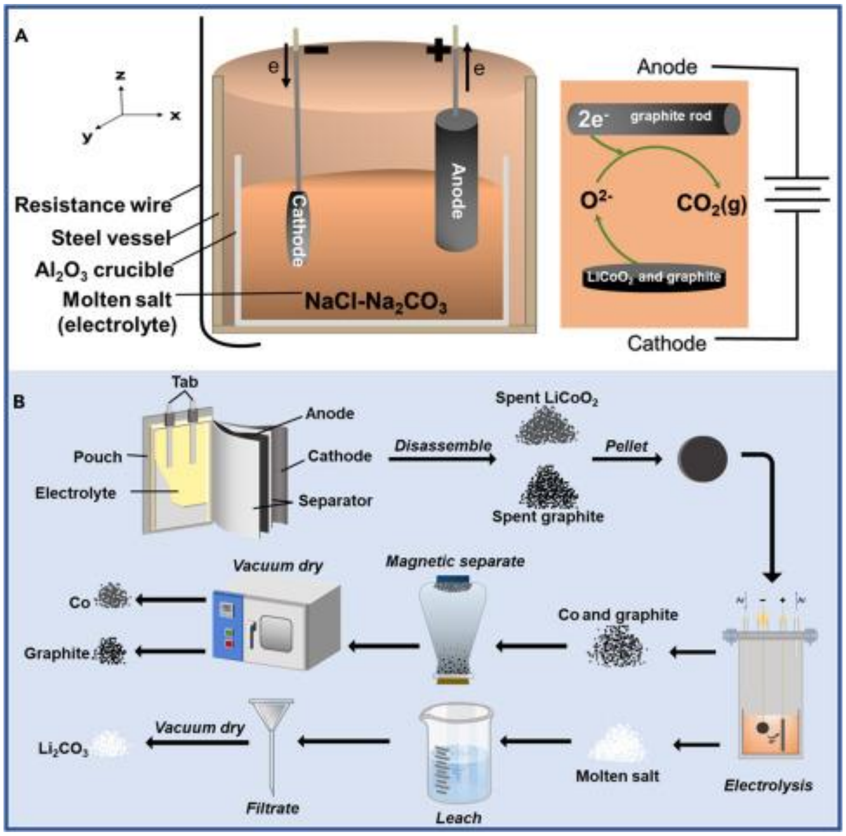

Figure 8. (a) Experimental setup and (b) flow chart for the recovery of Co, Li, and graphite from spent $LiCoO_2$ batteries (Feng et al., 2023; open access).

4. Conclusion

This review summarizes the current procedures and technologies used in the recycling of spent lithium-ion batteries, which are widely used in a variety of applications such as consumer electronics, electric vehicles, and energy storage systems. The goal of ongoing research and development is to increase the cathode material recycling procedures scalability, cost-effectiveness, and efficiency. In order to drastically reduce the need for new raw materials and minimize the impact on the environment, it is intended to establish a continuously system where materials from mortality batteries are continuously recycled into new batteries. Overall, this review emphasizes the necessity of a multimodal strategy that builds on the advantages of different recycling techniques to develop a more extensive and long-lasting battery recycling infrastructure. In order to improve the effectiveness and security of recycling operations, future research should concentrate on improving process technologies, creating greener leaching agents and solvents, and incorporating automated systems. In addition, in order to promote the use of cutting-edge recycling technologies and facilitate the shift to a battery circular economy, policy frameworks and financial incentives are essential. Through the resolution of these obstacles and possibilities, the recycling sector can guarantee a sustainable supply of vital materials for the upcoming generation of energy storage technologies while also considerably mitigating the environmental impact of battery waste.

Conflict of Interests
The authors declare no conflict of interest.

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