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Unlocking the Potential of Electrolyte Recovery from Battery Waste: A Promising Path to Sustainable and Safe Li-Ion Batteries

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The growing use of rechargeable batteries, especially lithium-ion batteries, has created new challenges in battery waste management. The recycling of waste batteries has garnered significant interest worldwide as a means of alleviating resource constraints and mitigating the harmful environmental impacts. Recycling resources from spent batteries can help in establishing a sustainable value chain for materials required for battery production. While metal recovery has been widely studied, electrolyte recovery remains underexplored despite its hazardous nature. Electrolytes contain flammable and toxic components, such as lithium hexafluorophosphate. Electrolyte recovery presents challenges, including decomposition and evaporation during the recycling process. It can produce various harmful components, such as hydrogen fluoride and phosphoryl fluoride, which can be released into the exhaust gas stream. Electrolyte recovery can help prevent hazardous chemicals from being released into the environment. Electrolyte recovery will not only contribute to resource conservation but also pollution reduction. This review examines current strategies for electrolyte recovery from spent batteries, evaluating their benefits and drawbacks, as well as the challenges and potential solutions for more effective implementation.

1. Introduction

The world is transitioning to electric modes of transportation, which helps reduce carbon emissions, and the backbone of battery technologies has become a focal area of development.[1,2] Lithium-ion batteries (LIBs) are the dominant battery technology and have become an integral part of day-to-day life.[3] Their ubiquity is achieved through our electric vehicles (EVs), phones, and laptops, which are used for power and a sustainable lifestyle.[4,5] A typical commercial LIB consists of four major components: a cathode, a separator, an electrolyte, and an anode.[5] The electrolyte plays a vital role in determining the performance of a LIB, where Li-ions carrying electrons move back and forth between the electrodes (cathode and anode).[1,6] The urgent need for batteries with higher energy density and safety leads to the development of high-capacity cathode materials, continuously increasing the stability requirements of the electrolyte.[1,7,8] The ideal electrolyte should have the following characteristics:[1] 1) electrochemical stability window and can form stable electrode/electrolyte interphases, 2) higher ionic conductivity (more than $10\,mS\,cm^{-1}$), 3) lower electronic conductivity, 4) large electrochemical stability window, 5) high wettability of the electrode's surface, and 6) environmentally friendly.[1,9,10]

The primary goal for batteries is to be produced sustainably, with a low carbon footprint and an optimized circular economic approach throughout all steps of the value chain.[11] Indeed, sustainable battery technology is strongly supported worldwide. To implement a circular economic approach to battery technology, active research on sourcing, extraction, and processing of raw materials is crucial, along with a focus on establishing a materials traceability database.[11] In the context of the growing battery industry and supply, research on recycling technologies is highly demanded.[11] Increasingly, LIBs are powering EVs, as evidenced by the several million battery EVs already on the road. The skyrocketing of EV production has already pledged to ban sales of fossil-powered vehicles by 2040–2050.[12] The spent batteries are expected to generate 2 million metric tons per year by 2030. And yet, there is no clear idea of what is to happen to these spent batteries after their lifetime.[13,14] Currently, fewer than 5% of LIBs worldwide are recycled, while the rest end up in landfills. Most recently, the number of spent batteries has significantly increased,[15] presenting new challenges for waste management in the recycling industry.[15] The recycling of waste batteries has

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gained enormous interest globally to alleviate resource shortages and mitigate the detrimental environmental impact caused by spent battery waste.[15,16] With the inevitable increase in EV production over the coming years, recycling batteries is key to the future.[17] The recycling process facilitates the reuse and reduction of raw material shortages in a circular economy.[18] The EV market is expected to grow, necessitating the development of circular economy designs for end-of-life (EOL) products and the recovery of materials for secondary batteries.[17] By bridging the gap between academic and industrial research approaches, recycling electrolytes will provide critical insights into the potential of future batteries and sustainability.[17] As in most studies, metal recovery has gained significant attention, while electrolyte recovery remains underexplored. Hence, this review focuses on electrolyte salt recovery from spent battery waste, as this aspect will help in understanding electrolyte waste management and provide information about recycling electrolytes. This comprehensive review examines various strategies for recovering electrolytes from spent battery waste, including processes for recycling electrolytes. It highlights the challenges and potential solutions for implementing a more effective electrolyte recovery strategy from spent battery waste.

2. Electrolyte Composition in Li-Ion Batteries

The electrolyte is one of the four major components in batteries, playing a key role in transporting Li-ions during the cycling process, and is often referred to as the "heart" of LIBs.[9] The electrolyte is a crucial component and is composed of lithium salt (lithium hexafluorophosphate ($LiPF_6$)) and is dissolved in organic carbonated solvents.[19] Solvents such as ethylene carbonate (EC), dimethyl carbonate (DMC), propylene carbonate (PC), and ethyl methyl carbonate (EMC) facilitate the migration of lithium ions throughout the battery's charging and discharging cycles for LIB (**Figure 1**).[20–23] The electrolyte utilizes different lithium salts (e.g., $LiPF_6$, $LiCF_3SO_3$, $LiBF_4$, $LiAsF_6$, $LiClO_4$, $LiPO_2F_2$, and $Li(SO_2CF_3)_2$) in an organic solvent.[24] On top of the mentioned salts, $LiAsF_6$ and

$LiPF_6$ have the highest ionic conductivity, whereas $LiClO_4$, $LiBF_4$, and $LiCF_3SO_3$ are lower. $LiPF_6$ is readily decomposed into PF_5 and LiF, and the preparation of salt is complicated and expensive. However, $LiAsF_6$ poses challenges in terms of decomposition during disposal and toxicity. The strong oxidizing property of $LiClO_4$ poses significant safety concerns. Additionally, additives such as vinylene carbonate, fluoroethylene carbonate, and LIBOB are used in electrolyte mixtures, among others. The solvent and lithium salt contribute to the ionic conductivity and stability of the electrolytes. Additives can also play a role in forming solid electrolyte interphase (SEI), which has high ionic conductivity, and helps suppress the detrimental growth of SEI. Ions, such as Li^+, are dissolved in the electrolyte and carry out charge transfer reactions, including reduction or oxidation, at the electrode surface.[25] The recovery of electrolytes from spent batteries is not only beneficial for environmental protection and economic effectiveness but also leads to the creation of high-value products from these recovered materials. The cost of electrolytes is very expensive due to the use of Li salts, which is a more critical factor to address for future sustainable batteries.

3. Current Scenario for Electrolyte Recovery from Spent Battery Waste

Most research on spent LIB recycling focuses on recovering valuable metals like cobalt, lithium, nickel, and copper. However, due to its volatile, flammable, and toxic nature, the aged electrolyte must first be removed during this process. Additionally, the wide variation in electrolyte compositions across different LIBs increases the complexity and cost of recycling.[26,27] The increasing volume of waste electrolytes makes high-value recovery and reuse more crucial than ever for environmental protection and resource conservation. Efficient and environmentally friendly recycling of electrolytes is vital for achieving carbon neutrality. Clean recovery processes can help address resource shortages, support circular economic initiatives, and reduce environmental and health risks.[28] This section provides an overview of current
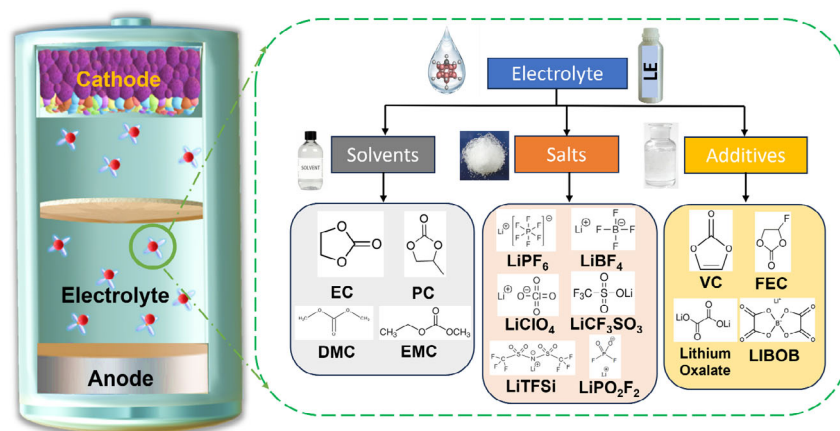
Figure 1. A schematic of LIB and electrolyte components. A liquid electrolyte consists of solvents, salts, and additives.

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electrolyte recovery methods, challenges, and future directions. From a broader perspective, encompassing environmental, economic development, and carbon neutrality, the article underscores the urgent need and critical role of electrolyte recycling in advancing green innovation within the LIB industry.

3.1. Possible Routes for Electrolyte Recovery or Treatment: Electrolyte Recovery Approaches

LIBs contain organic materials such as electrolytes and binders. Most commonly used binders are polyvinylidene fluoride (PVDF) and carboxymethyl cellulose (CMC) to help secure the active materials to the current collectors.[29] Different strategies are studied for the recovery or treatment of spent battery electrolytes. The composition of the electrolyte, safety considerations, and economic viability influence the choice of method. The different recycling methods include thermal decomposition (Pyrolysis), solvent extraction, supercritical CO_2 extraction, and other methods such as mechanical separation, alkaline absorption, and freezing.

3.1.1. Thermal Decomposition Method

In the thermal decomposition process, high temperatures in an inert atmosphere are used to decompose organic solvents into low-molecular-weight compounds. The advantages of the pyrolysis process are ability to prevent the oxidation of valuable metals, thus allowing for the efficient recovery of cobalt and lithium. Furthermore, it facilitates the safe disposal of $LiPF_6$ without causing environmental harm.[30] Research on the pyrolysis of spent LIBs mainly focuses on three areas: 1) understanding the reaction kinetics of the pyrolysis process, 2) developing separation techniques that make use of pyrolysis, and 3) identifying and tracking the toxic gases released during the process.[31] Kinetic studies show that the electrolyte, separator, and binder in spent LIBs begin to volatilize and break down between 20 and 500 °C.[31] In this temperature range, more than 96% of the electrolyte, about 88% of the separator, and around half of the PVDF binder are transformed into pyrolytic gases and oils. These products primarily consist of short-chain hydrocarbons and hydrofluoric acid (HF), which can be effectively neutralized using alkaline solutions.[32] Multistage pyrolysis technology has been developed based on the varying decomposition temperatures of different organic components. This approach enables the environmentally safe treatment of electrolytes and allows for the efficient separation of electrode materials.[33] To minimize the release of harmful gases and increase the recovery of cathode materials, vacuum pyrolysis has been receiving increasing attention.[31] Researchers have extensively studied how pyrolysis temperature and vacuum level influence the separation of cathode sheets and the phase transformation of valuable metals in the active material cathode powder.[34] Using the "metallurgy first and separation later" approach, organic components like the separator and binder are converted into pyrolytic oil and gas. At the same time, the cathode active material is reduced, enabling the extraction of valuable metals via acid leaching without additional reducing agents (**Figure 2**a).[35] Researchers revealed that interactions during pyrolysis, such as PVDF forming a liquid film and active materials adsorbing electrolyte residues, impede the complete removal of organics, leading to residual fluorine contamination. Higher pyrolysis temperatures help remove organics but also increase fluorine retention in the form of metal and organic
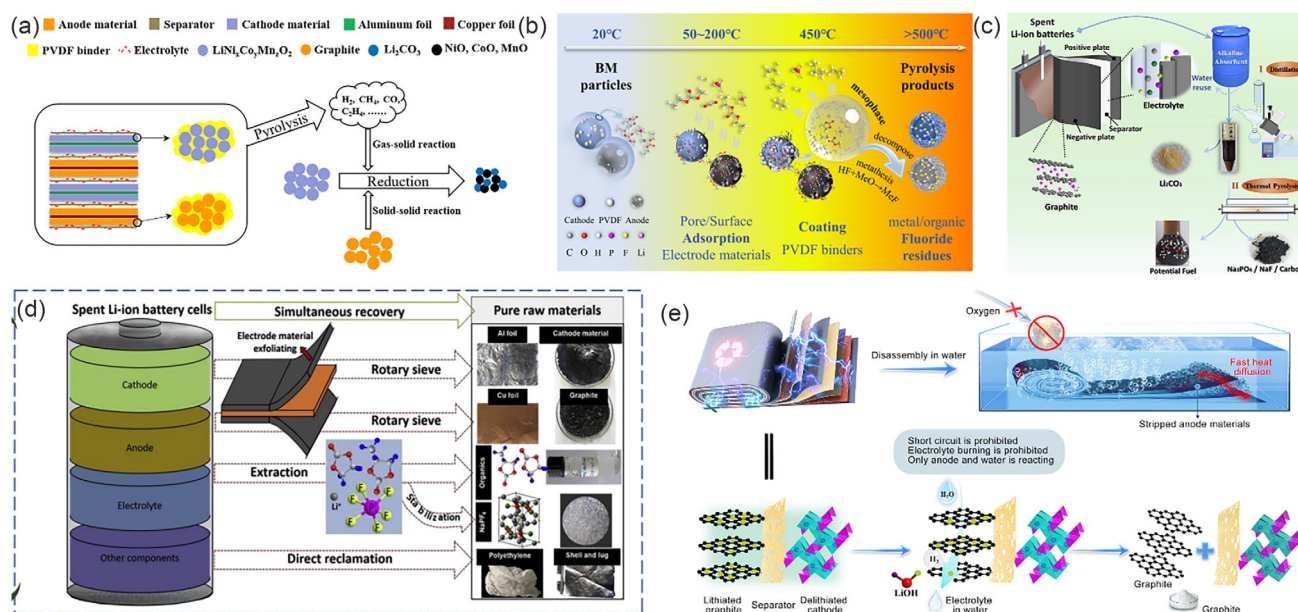
Figure 2. a) Schematic illustration of the pyrolysis and reduction process of spent LIB components. Reprinted with permission.[35] Copyright 2021, American Chemical Society. b) Possible transformation pathway of fluorine-based contaminants during the pyrolysis of BM. Reprinted with permission.[31] Copyright 2022, Elsevier. c) Two-stage heat treatment process flow diagram for waste electrolyte liquid. Reprinted with permission.[30] Copyright 2024, Elsevier. d) Schematic diagram of recovered electrode materials and hazardous electrolytes. Reprinted with permission.[40] Copyright 2019, Elsevier. e) An illustration of the dismantling of spent LIBs in water. Reprinted with permission.[41] Copyright 2022, Elsevier.

fluorides. The findings offer insights into optimizing clean pyrolysis processes to achieve more effective defluorination of black mass (BM) materials.[31] As described in Figure 2b, during BM pyrolysis, fluorine-containing pollutants transform in three stages: 1) 20–200 °C: electrolyte decomposes, releasing fluorinated gases and leaving residues due to adsorption; 2) 200–400 °C: secondary pyrolysis of residues and SEI decomposition occurs; 3) 400–500 °C: PVDF melts into a liquid phase, promoting reactions that embed fluorine in electrode materials. These residues reduce BM purity and cause corrosive leachates.[31] A low-temperature thermal treatment ranging from 90 to 150 °C demonstrated potential for recovering electrolyte solvents from spent LIBs ahead of metal recycling. Organic solvents, such as DMC, EMC, and EC, were successfully reclaimed, with EC primarily obtained in a solid form. The best recovery was achieved at 130 °C in 80 min. Harmful gases like HF and phosphorus oxyfluoride (POF_3) were effectively removed using water-based gas scrubbing, resulting in the formation of hydrofluoric and phosphoric acids as byproducts. Further research is needed to improve solvent recovery efficiency and evaluate the reusability of the recovered materials.[36] The electrolyte system in waste LIBs is complex, and its transformations during pyrolysis are multifaceted. Thus, a two-stage thermal treatment was employed to investigate the transformation of residual electrolytes in spent LIBs, as illustrated in Figure 2c. During distillation, lithium formed lithium carbonate, while the hexafluorophosphate remained settled without undergoing hydrolysis. Pyrolysis converted most of the concentrated electrolyte (76.21%) into valuable oil and 8.89% into gases, such as CO and CO_2, with 300 °C as the optimal temperature. Hexafluorophosphate was stabilized as fluoride and phosphate, and the addition of KOH helped immobilize these, thereby reducing corrosion risks. The process effectively recovers useful products, minimizes environmental impact, and shows strong industrial application potential.[30] Pyrolysis is a simple method with a high processing capacity that enables recycling electrolytes and binders. However, it produces harmful gases like fluorides, requiring specialized treatment systems. Another issue in this process is the need for high equipment investment and operational costs.

3.1.2. Organic Solvent Extraction

Solvent extraction is a versatile technique that enables control over product composition and the recovery of pure materials. Traditional aqueous solvent extraction involves transferring target components from the aqueous phase to an organic phase through the formation of a complex. Additionally, nonaqueous phase extraction uses deep eutectic solvents to extract metal ions from low-water-content leaching solutions.[37] The electrolyte in LIBs can be dissolved in an organic solvent and then recovered by distillation based on the solvent's boiling point. For example, the solvent extraction process has been described in a Chinese patent.[38] In this method, spent LIBs are first dismantled to remove the battery core, which is then immersed in carbonate solvents (such as EC, PC, DEC, (EMC) ethyl methyl carbonate or mixtures).[39] After immersion, the materials undergo filtration, centrifugation, treatment with anhydrous HF gas, and vacuum distillation at 100 °C to remove solvents from the electrolyte, extractants, and high-purity $LiPF_6$.[38] Another study developed an environmentally friendly process to recover electrode materials and hazardous electrolytes from LIBs without using strong acids or alkalis. This allowed the electrode materials to be reclaimed in a flaky form.[40] Figure 2d illustrates a schematic diagram of the recovered electrolyte. A novel process was developed to simultaneously peel off electrode materials from metal foils and recover the electrolyte in a controlled and environmentally friendly manner. The researchers first analyzed the adhesive forces between Al foil and the cathode coating, both theoretically and experimentally. They determined the minimum amount of Al that needed to be dissolved to overcome these forces. Based on this analysis, an aqueous solution called aqueous exfoliating and extracting solution (AEES) was formulated to precisely control the dissolution of the Al foil, thereby enabling the effective separation of cathode material. AEES, composed mainly of sodium salts and surfactants, functions by partially dissolving the PVDF binder and weakening the mechanical and electrostatic forces that hold the active materials to the foil. Additionally, AEES was able to dissolve the anode binder (CMC), allowing the graphite to be cleanly separated from copper foil. The solution also served as a substitute for conventional organic solvents in electrolyte recovery: it extracted EC and PC, which were subsequently recovered through distillation. Simultaneously, stabilized $LiPF_6$ was precipitated and recovered by filtration. The study further evaluated the efficiencies of electrode material separation and electrolyte recovery, along with an analysis of the overall recycling cost. By optimizing the AEES concentration, the process achieved high recovery efficiencies of 95.6% for the electrolyte. Studies have shown that disassembling batteries in an aqueous medium can effectively absorb the electrolyte and reduce gas emissions.[41] After carefully separating spent LIBs in water, a solution containing lithium, electrolyte, and water-soluble binders can be obtained.[42] At the same time, the soluble lithium salts and electrolyte dissolved in the water can be further processed for recovery, minimizing lithium loss and reducing fluoride emissions.[42] The recycling mechanism is illustrated in Figure 2e.[41] Solvent extraction can recover electrolytes from used LIBs, but the loss of extraction agents increases costs and causes pollution. The recovered electrolyte contains impurities from solvents, raising purification costs and reducing performance in new LIBs. Despite avoiding harmful byproducts, the large volume of solvents can lead to contamination and higher reuse costs.

3.1.3. Supercritical CO_2 Extraction

Due to their low surface tension and viscosity, supercritical fluids can penetrate solid matrices that regular liquids cannot, thanks to their gas-like viscosity and liquid-like density.[42] Near the critical point, the pressure and temperature of supercritical fluids can be quickly adjusted to extract and separate components based on their solubility. With their unique properties, supercritical fluids are ideal for extracting electrolytes from spent LIBs. CO_2 is the

most promising medium due to its achievable critical point, high dissolvability of nonpolar solvents, efficient extraction, and non-toxic, pollution-free nature.[43] Supercritical CO_2 extraction was used to recover organic carbonate-based electrolytes from spent LIBs. Using response surface methodology and dynamic extraction, a yield of 85.07% was achieved. The extracts were analyzed, confirming that supercritical fluid extraction is an effective method for electrolyte recovery, helping to reduce environmental pollution and resource waste.[44] Grützke et al.[45] used an autoclave extraction method, employing supercritical carbon dioxide under high pressure, to recover electrolytes from different separators and commercial LIBs. The results showed that polyethylene fleece could recover $73.5 \pm 3.6\%$ of the electrolyte, while glass fiber recovered only $36.7 \pm 1.6\%$. The method was optimized to address the lower recovery rates using supercritical and liquid CO_2 with different solvents. Liquid CO_2 proved more effective for extracting linear carbonates (DMC, EMC), while supercritical CO_2 was better for EC.[42,45] Liu et al. demonstrated that leveraging properties such as polarity and melting point can enhance the separation and recovery of carbonate-based electrolytes, thereby improving their recyclability. The influence of the physical properties of carbonates on their behavior during supercritical CO_2 extraction was studied under specific conditions using gas chromatography with flame ionization detection.[46] Results showed that polarity plays a crucial role in the extraction process: nonpolar carbonates are most effectively extracted using nonpolar or weakly polar media, which reduces energy consumption and maintains high yields, whereas polar carbonates require polar solvents or cosolvents. During collection, both polarity and volatility affect the order of separation, with polar carbonates being collected before nonpolar ones.[47] Supercritical CO_2 extraction, combined with resin and molecular sieve purification and the addition of electrolyte components, affected the recycled electrolyte (**Figure 3**a).[48] The recovered electrolyte showed an impressive ionic conductivity (0.19 mS cm^{-1} at 20 °C), on par with commercial options, and remained stable up to 5.4 V. When tested in a Li/LiCoO$_2$ cell, it delivered an initial discharge capacity of 115 mAh g^{-1} and, after 100 cycles at the current rate of 0.2C, maintains the capacity retention of 66%.[48,49] These results highlight its strong potential for reuse and represent a meaningful step forward in electrolyte recycling. Mu et al.[50] reported that the exfoliation process of cathode electrodes in supercritical CO_2 is illustrated in Figure 3b. Supercritical CO_2 extracted the electrolyte and binder at high temperature and pressure, weakening the bond between the cathode materials and the aluminum foil.[51] When the pressure is released, the reduced adhesion cannot withstand internal stress, resulting in the clean separation of the cathode material in sheet form while maintaining the integrity of the aluminum foil. Zachmann et al. investigated the use of sub- and supercritical CO_2 to selectively recycle the electrolyte from spent LIBs, aiming to reduce hazardous emissions and safety risks associated with traditional high-temperature recycling methods. Their study demonstrated that nonpolar solvents, such as DMC and EMC, could be selectively recovered, while minimizing toxic gas release and preserving LiPF$_6$ integrity.[52] Currently, supercritical CO_2 extraction is used to recover electrolyte solvents from spent LIBs. Studies on EC solubility under various temperatures and pressures revealed an increase in solubility with higher pressure. The Chrastil model was used to fit the data. Proof-of-concept experiments confirmed that EC can be efficiently extracted at 140 bar and 40 °C without altering its crystal structure.[53] Another study reported that supercritical CO_2 extraction is an effective alternative to conventional methods, such as leaching and pyrolysis, for removing the PVDF binder from secondary batteries. The results indicated that a high binder removal efficiency of 98.8% was achieved under optimal conditions with dimethylformamide (DMF) as a cosolvent.[2] Notably, the type of solvent used significantly impacted battery performance, with cycling stability improving in the order of DMF,[2] *N*-methyl-2-pyrrolidone (NMP),[17,54] dimethyl sulfoxide (DMSO), and dimethylacetamide (DMA).[2,55] Supercritical CO_2 extraction has emerged as a selective, environmentally friendly, and safe method for electrolyte recovery, avoiding solvent-derived impurities and overcoming polarity-related limitations of target compounds. However, its effectiveness is limited by the inability to extract lithium salts, such as LiPF$_6$, without the aid of additional organic solvents. It also requires precise control of temperature and pressure, making industrial scalability and cost-effectiveness key challenges for widespread application.
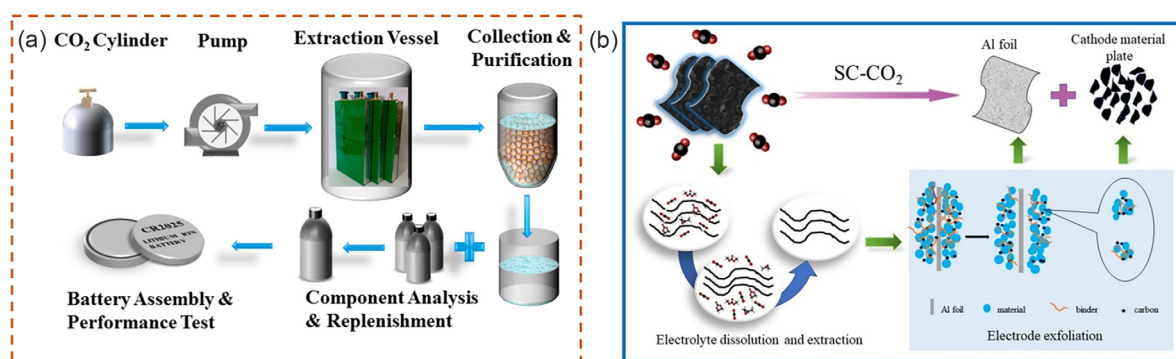
Figure 3. a) Schematic approach of the electrolytes recovered from spent LIBs. Reprinted with permission.[48] Copyright 2017, American Chemical Society. b) Proposed exfoliation mechanism of the cathode electrode in supercritical CO_2 fluid. Reprinted with permission.[50] Copyright 2022, Wiley.

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3.1.4. Other Approaches

Electrolytes are removed from dismantled LIBs using mechanical methods such as centrifugation, air purging, and vacuum filtration under inert atmosphere conditions.[39] For example, Li et al.[56] developed a method to recover electrolytes from waste LIBs by first disassembling the batteries and using centrifugation to extract the electrolyte. The recovered electrolyte is then purified through filtration, decolorization, and dehydration. After analyzing its composition, additional components are added to match standard electrolyte formulations, making it suitable for reuse in battery production.[56] To recover electrolyte from waste LIBs, the process involves puncturing the shell, collecting the outflowing electrolyte, and utilizing pneumatic pressure to extract the remaining electrolyte. The recovered electrolyte is then treated with a barium oxide solution to recover lithium fluoride, facilitating efficient recycling of both the electrolyte and lithium.[57] Xiao et al. highlighted how organic solvents and fluorinated compounds migrate and transform during mechanical treatment of LIBs, simulating real recycling conditions. LIB components were categorized into three forms: strip-shaped, powdery, and liquid. Mechanical processing converts strip-shaped materials into large or mid-sized particles and powdery materials into fine particles. Notably, liquid electrolytes underwent significant deformation, with residues mainly found in plastics, anodes, and cathodes. Volatile organic solvents were released into the air, posing fire, and health hazards, while $LiPF_6$ decomposition products presented risks of corrosion and toxicity. These insights help guide safer handling of hazardous substances in battery recycling.[58] Generally, mechanical methods cannot fully extract electrolyte due to its absorption in electrodes, separators, and SEI layers. Therefore, solvent washing is often necessary in conjunction with mechanical processes. All operations must be carried out under an inert gas atmosphere to prevent the electrolyte from reacting with moisture in the air.[39]

In the freezing method, electrolytes are recovered by converting them into a solid, thereby preventing evaporation and decomposition, and facilitating easier collection. Alternatively, distillation can be used by heating spent LIBs to evaporate the electrolyte and then recondensing it into a liquid. Both approaches rely on phase transitions between solid, liquid, and gas states. For instance, Zhao et al. demonstrated that volatilizing the electrolyte, utilizing low-temperature freezing to mitigate its harm, and adding water during distillation to facilitate the decomposition of $LiPF_6$ enabled the collection of the electrolyte without damaging the battery core, thereby reducing the need for recycling battery materials.[59] Distillation and freezing methods can efficiently collect electrolytes by controlling phase changes. Distillation collects solvents but decomposes $LiPF_6$, generating LiF and requiring equipment to handle toxic byproducts. Freezing reduces electrolyte activity and prevents the release of harmful substances, although its high cost and equipment requirements limit its use.

Alkaline adsorption can be utilized in a harmless treatment process for recovering waste LIB electrolyte. For example, Cui et al. reported a process that began by sorting and crushing batteries after cooling them at low temperatures with liquid nitrogen. The batteries were then treated in a three-stage alkali

process using a calcium hydroxide ($Ca(OH)_2$) solution. After the third-stage treatment, the liquid was discharged into a coagulation tank, where it was further treated with an inorganic salt coagulant. The tail gas was treated harmlessly by water spraying. The system included a sorter, hopper, low-temperature processor, closed shear crusher, three-stage reaction tank, coagulation pool, and spray tower, all connected in series. The method was simple, environmentally friendly, economical, and effective, with the alkaline solvent reusable after deployment.[60] NaOH was used to soak disassembled spent LIBs, dissolving soluble components. After filtration, xylene was added to the filtrate to extract organic compounds. The organic phase was distilled to recover dimethoxyethane, and the remaining aqueous solution was adjusted to a basic pH before adding sodium carbonate to precipitate crude lithium carbonate.[61] However, this alkaline absorption process produces many impurity ions, generates large volumes of high-pH wastewater, and offers limited recovery efficiency, making it costly and less sustainable.

3.2. Challenges in Electrolyte Recovery for LIBs

Although recycling electrolytes from spent LIBs is essential, several challenges remain in developing effective technologies for industry: 1) Pyrolysis provides a simple, high-throughput method for recovering electrolytes and adhesives. However, it generates hazardous gases, such as fluoride, which require advanced purification systems and involve considerable equipment and operating costs. 2) Solvent extraction can recover LIB electrolytes, but challenges include high solvent consumption, contamination risks, and increased purification costs, which reduce the quality and cost-effectiveness of the recovered materials. 3) Although advanced extraction methods such as supercritical CO_2 extraction show promise, issues remain regarding recovery efficiency, product purity, and environmental impact, which impede large-scale industrial applications. Cost and performance parity with virgin electrolytes also remain significant barriers for practical regeneration. 4) Challenges in electrolyte recovery for LIBs include the complex and unfixed composition of electrolytes due to proprietary formulations and ongoing development of new electrolytes, making it difficult to develop universal recycling processes. 5) Additionally, the highly volatile, flammable, toxic, and sensitive nature of electrolytes complicates collection and handling, especially during the continuous cycling process, where electrolyte invalidation occurs due to internal reactions and external heat-induced decomposition. The mixing of spent batteries hinders targeted recovery, while environmental concerns related to hazardous byproducts, such as HF and HCl, pose additional challenges.

Considering all the above points, the main challenges in electrolyte recovery and recycling are hazardous EOL materials, high costs, and lack of standardization.[62] When LIBs reach the end of their life, their performance declines due to various aging processes that affect the battery function and lead to the creation of harmful and corrosive substances inside the cells. $LiPF_6$, a common LIB electrolyte salt, becomes unstable when mixed with

solvents and exposed to battery operating conditions, especially moisture, which leads to the formation of toxic HF.[63] Grutzke et al. found that during long-term storage of shredded battery materials, harmful organofluorophosphate compounds can form. Although initially present in low amounts, these toxins could become dangerous at higher temperatures and in large-scale recycling environments.[64]

Solvent recovery from LIBs is effective; however, varying chemical properties lead to inconsistent results. Recycling $LiPF_6$ is more difficult due to its volatility and potential to cause safety and contamination issues. The lack of standardization in electrolyte composition complicates continuous recycling. Economically, electrolytes account for $\approx 13\%$ of the cost of a new battery; however, their value drops significantly once the battery reaches the EOL. Environmentally, continued use of conventional disposal methods such as incineration will lead to a significant increase in greenhouse gas emissions. If current trends continue, CO_2 emissions from incinerated electrolytes could reach 90,000 tonnes by 2030. Even with increased recycling rates, emissions will rise unless more sustainable disposal methods are introduced.[65]

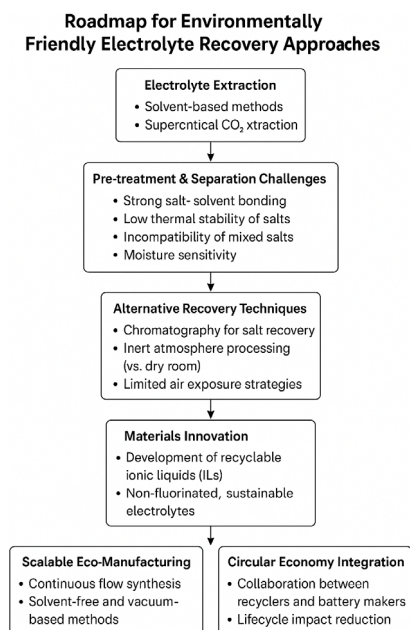### 3.3. Roadmap for Environmentally Friendly Electrolyte Recovery Approaches

After electrolyte extraction, whether through solvent-based methods or using supercritical CO_2, it must be further processed into a product that can be resold. Ideally, each component of the electrolyte should be separated; however, this involves many complex steps, making it economically impractical. A significant challenge is the strong bond between the salt and solvent, which results in low thermal stability and renders simple recovery methods like evaporation ineffective.[62] More cost-effective techniques, such as chromatography, may offer a solution for extracting and reusing salt.[66] Utilizing new lithium salts in electrolytes to enhance battery performance. However, the newer salts are difficult to separate cost-effectively, and if separation isn't possible, the resulting mixed salts may have little commercial value. Reuse might only be feasible through manufacturer-specific recycling processes. However, as battery technologies advance, the types of electrolytes may change, making the recovered salts incompatible with future battery designs. As a result, their reuse may be limited to serving as basic chemical raw materials rather than being directly used in new batteries.[67] In addition to separation difficulties, lithium salts are sensitive to moisture, which makes recycling more challenging, as the recovery process in a dry room significantly raises costs and energy consumption. A better way of the breakdown kinetics is essential for developing scalable and efficient processes, particularly to ensure a low acid content in the recovered electrolyte. Studying how other battery components may trap harmful byproducts like HF during recycling is also essential. Alternatives to fully dry room environments include using inert atmospheres or limiting exposure time to air, though practical stability testing is complex and depends heavily on scale and equipment.[63]

Direct recycling of electrolytes into new batteries is not feasible, so alternative methods, such as concentrating spent electrolytes and recovering solvents for salt production, are necessary. Recycling is complicated by toxic degradation products, unstable salts, and inconsistent formulations, which make it challenging to separate salts from solvents. Although some companies have successfully recovered electrolytes for resale, widespread recycling remains uncommon. Future advancements depend on collaboration to develop more recyclable electrolytes that promote a circular economy and reduce environmental impact.[62] There is an increasing focus on the development of cost-effective, sustainable, and nonfluorinated chemical electrolytes, such as ionic liquids(ILs).[68] This includes not only the development of greener chemical formulations but also the advancement of scalable and environmentally friendly recovery and production processes. These include: 1) Designing ILs that are easier to recover and recycle. 2) Developing continuous flow synthesis techniques to reduce waste and energy use. 3) Innovating processing methods that align with green chemistry principles (e.g., solvent-free methods, vacuum-based processing). 4) Exploring biobased chemicals, such as biomass-derived organic acids or green solvents, to reduce the dependency on hazardous solvents. These efforts aim to reduce environmental impact across the full lifecycle of electrolytes, from production to recovery and reuse. The roadmap for environmentally friendly electrolyte recovery approaches is depicted in **Figure 4**.

4. Safety, Techno-Economic Aspects of Electrolyte Recovery, Cost Perspective

In this section, the battery safety, cost, and techno-economic challenges associated with the electrolyte recovery process are discussed. Nevertheless, the electrolytes raise vigorous environmental

Figure 4. Roadmap for environmentally friendly electrolyte recovery approaches.

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and safety concerns that can't be ignored, which might be attributed to the highly volatile and flammable nature of organic solvents.[69,70] Furthermore, these organic solvents can break down into harmful byproducts, especially when mechanically broken down, overcharged, or exposed to elevated temperatures over an extended period. This is one of the crucial concerns when it comes to battery safety and the problems associated with recycling them.[70] The breakdown of electrolyte components can lead to hazardous conditions, such as thermal runaway, which poses risks to the batteries, the individuals using them, and the surrounding environment. These factors underscore the importance of conducting further research on safer electrolyte formulations and improved battery management systems, enabling LIBs to be safer and last longer in an increasingly electrified environment.

4.1. Battery Safety Risks Associated with Electrolytes

LIB electrolytes can readily catch fire and become unstable when they become too hot or undergo exothermic reactions, which poses a significant safety risk, mainly due to the thermal runaway phenomenon.[71] This self-sustaining exothermic process can lead to disastrous outcomes, such as fires or explosions, which are highly hazardous to people and property.[72] For instance, at high temperatures, the breakdown of $LiPF_6$ under wet conditions results in the production of hazardous HF and POF_3. These byproducts, such as POF_3 and HF, are not only highly corrosive but also cause a profound impact on human health and can damage industrial equipment.[73] For instance, in a recent study, Zachmann and his colleagues investigated electrolyte recovery from spent LIBs and reported their findings elsewhere.[36] They found that electrolyte solvents, such as EMC, DMC, and EC, could be successfully recovered within 80 min at a temperature of 130 °C. However, $LiPF_6$ could produce LiF and partially decompose at a temperature of 130 °C. In contrast, above 150 °C, it fully decomposed, with corresponding byproducts POF_3 and HF formed (**Figure 5**a).

Furthermore, the plot reveals that at low temperatures, decomposition may be incomplete, resulting in a significant deviation in weight loss. In contrast, at high temperatures, the deviation is relatively low. One of the decomposition byproducts, POF_3, was found in the exhaust gas stream and confirmed by its characteristic peaks at different time intervals, as displayed in Figure 5b. The authors conclude that thermal treatment below 150 °C is a feasible process for solvent recovery before the metal recycling stage, offering feasibility, ease, and environmental safety.[36] This error deviation is significant since these emissions comprise a substantial portion of the release of harmful gases during necessary steps, including crushing, discharging, and thermal processing of used batteries. To overcome such difficulties, scientists have developed novel approaches for producing electrolytes, including the addition of flame-retardant chemicals with lithium salts (LiDFOB), which have high thermal stability, and the development of new methods to produce solid-state electrolytes. Very recently, Hsu and the team reported an innovative two-step method that involves antisolvent-assisted precipitation (ASP) and distillation to isolate and purify

both lithium salts and solvents from the spent electrolyte, as demonstrated in Figure 5c.[74] Using the ASP method, the authors selectively isolate $LiPF_6$ in the form of a complex with EC. The distillation process that follows removes the volatile carbonated solvents, allowing for the more effective recovery of $LiPF_6$ salts.

This innovative two-step process not only enables high-value $LiPF_6$ recovery and the use of carbonated solvent but also creates opportunities for environmentally friendly and circular recycling of LIBs. All of these are potential ways to mitigate the hazards associated with the spent recovery of electrolytes. Most commercial LIBs, on the other hand, still use regular liquid electrolytes.[75] This dependence highlights the need for the immediate implementation of robust safety rules, not only during the use of these batteries but also during their recycling operations to protect the environment.

4.2. Techno-Economic Aspects of Electrolyte Recovery

To date, various novel approaches have been implemented to recover the spent electrolyte in LIBs.[39,76] Each method has its individual economic and technical merits and demerits. 1) The simple mechanical techniques for recovering spent electrolytes include vacuum filtration, centrifugation, and purging with an inert gas.[58] These methods are viable, but they couldn't achieve 100% recovery of all the electrolyte.[77] Additionally, using excessive solvents for the electrolyte removal process reduces the overall efficiency of the process. 2) The combination of distillation cum freezing processes is another well-known approach to the recovery of spent electrolyte.[19,78] However, this process has its serious concerns, including the decomposition of $LiPF_6$, which produces harmful byproducts, as discussed earlier. The formed byproducts generate serious safety concerns, including an additional burden on instrument safety and further complicated environmental concerns. 3) On the other hand, the well-known solvent extraction process yields high lithium salts and a solvent recovery rate of over 90% from the spent electrolyte.[79] However, this process utilizes expensive solvents, and there is a chance of secondary pollution risks due to the vast amounts of solvents used, which should be handled carefully to minimize their environmental impact.[37] 4) CO_2 extraction process for spent electrolyte recovery is one of the most effective methods for extracting nonpolar liquids, offering excellent ecological benefits.[52] Still, it has inherent limitations, such as the need for cosolvents when working with $LiPF_6$. In addition, this process required a high-pressure instrument that can operate above 7 MPa, making it somewhat complicated. 5) Another efficient method for neutralizing fluoride species in the spent electrolyte is alkaline absorption, which involves extracting lithium salts in the form of lithium carbonate and lithium hydroxide.[49] However, this process generates a significant amount of alkaline wastewater, which requires high-cost remediation, making the process more complicated and challenging to sustain over time.

Therefore, new, integrated, and innovative processes must be conceived, such as low-temperature processes with mild chemical sequences involving precipitation, sublimation, and solvent-assisted extraction, which are capable of greatly minimizing
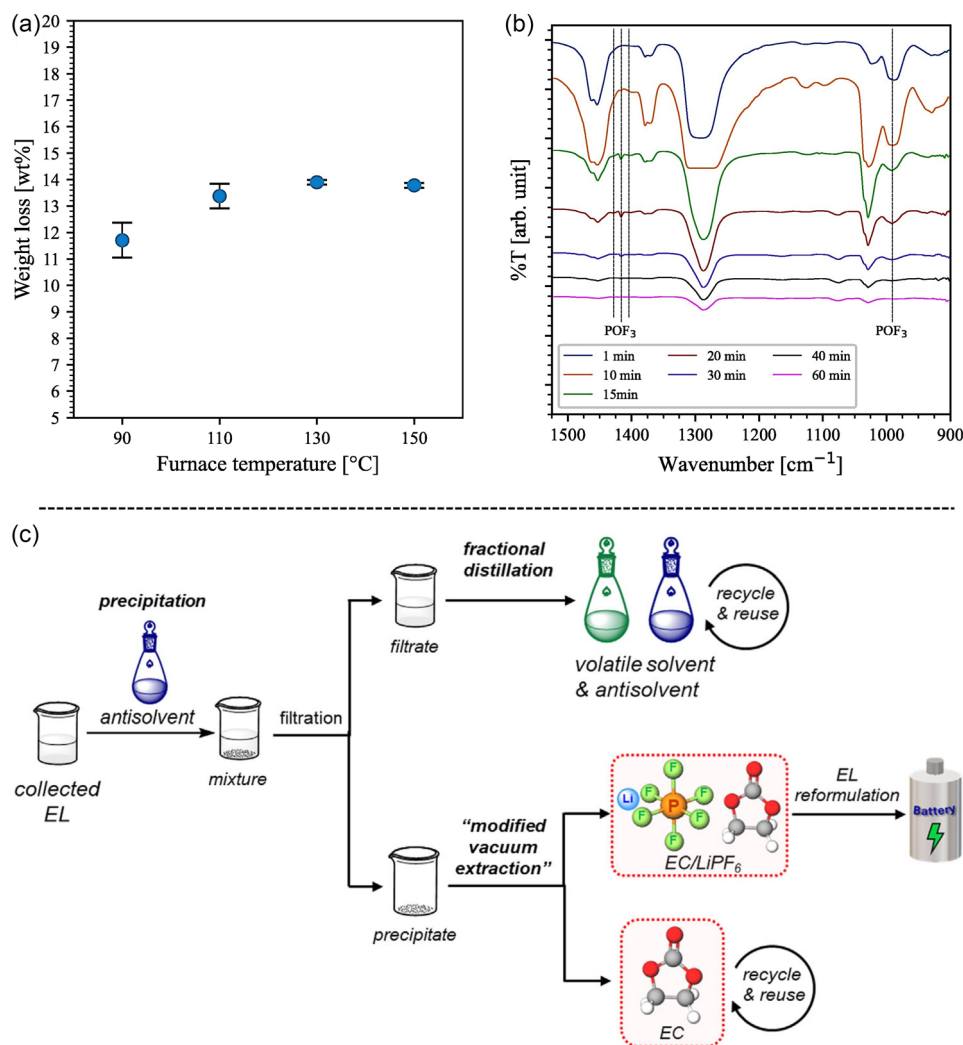
Figure 5. a) Thermal treatment of spent electrolytes at various temperature conditions for 3 h under an inert atmosphere and b) Fourier transform infrared spectrocopy (FTIR) results of exhaust gas recorded at various time intervals to confirm the formation of byproducts. Reprinted with permission.[36] Copyright 2023, Elsevier. c) A flowchart diagram illustrating a two-step electrolyte recovery process for isolating it from carbonated solvent EC. Reprinted with permission.[74] Copyright 2025, Wiley.

polluting gas releases. They also produce significantly higher recovery yields and lower energy use than existing high-temperature pyrolysis methods. Further implementing circular economy principles, particularly by recovering electrolytes before proceeding to the metal extraction steps, could be a decisive strategy. This proactive strategy not only strengthens the supply chain but also reduces the environmental damage associated with producing and recycling LIB electrolytes. These changes suggest a trend in the emergence of new, environmentally friendly, and high-performance technologies for recovering electrolytes from spent LIBs.

Despite numerous studies estimating resource recovery from spent battery waste, few studies have examined the economic cost of the electrolyte recovery approach separately. The composition of the feedstock (spent battery waste) and the technology adapted for recycling have a significant impact on the costs associated with recycling resources from spent battery waste. The cost of electrolyte recovery alone is not frequently reported, as during LIB

recycling, many other components, such as the cathode and anode, are also included with the electrolyte. For example, P. M. Tembo et al. reported that pyrometallurgy provides the lowest cost per tonne of LIB compared to hydrometallurgy process.[80] Furthermore, in another study, it was reported that the average recovery of electrolyte condensate from a ton of used and/or waste LIBs from electric cars is $\approx 12.5\%$. The estimated market value of the electrolyte condensate obtained during mechanical processing of (waste) electric car cells is ≈ 3480.5 USD ton^{-1}.[81] Additionally, the recycling costs of LIBs can range from \$2 to \$25 per kg, depending on the process and scalability.[82] It is evident that recovering organic solvents and lithium salts from the electrolyte will have a positive impact on the economy by reducing the need for lithium from primary sources. Additionally, the recovered organic components can be utilized in battery applications or for other purposes, such as material synthesis. This circular economy approach will encourage industries to recycle resources from the electrolyte, thereby generating economic benefits.

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4.3. Viable Industrial Approaches to Reduce the Cost Implications in LIBs

Several industrial solutions are under development to recover electrolytes, including various combinations of new techniques designed to improve safety, material recovery, and cost effectiveness, as well as driving added value from byproducts. Some of these strategies are:

4.3.1. Mechanical Separation and Distillation during Dismantling

By using machines, spent batteries or electrochemical cells will be dismantled into their constituent parts, and the distillation process will be used to purify and extract the valuable ingredients from the electrolyte resource.[83] The technique minimizes cross-contamination and maximizes yield by precisely controlling the detaching process.

4.3.2. Steam Treatment at Moderate Temperatures under an Inert Atmosphere

In this process, the materials being treated are maintained in a controlled atmosphere of inert gas to prevent reaction with oxygen from the air.[84] This process enables the rapid removal of volatile electrolyte components without degrading the materials, thereby maintaining quality for subsequent processing.

4.3.3. Catalytic Pyrolysis with Cathode Material Processing

This procedure utilizes catalytic pyrolysis, a thermal treatment that decomposes organic materials into simpler compounds by applying heat and a catalyst.[85] The catalytic agents facilitate the breakdown of complex electrolyte compounds, which aids in the extraction of simpler elements and the recycling of precious metals from cathode materials.

4.3.4. Mechanochemical Extraction with In Situ Catalysis during Crushing

This technology utilizes mechanical force to promote chemical reactions, allowing the extraction of the electrolyte as the materials are crushed.[86] Catalysis at this stage of synthesis significantly accelerates the reactions by breaking down the complex structures and facilitating the extraction of valuable compounds.

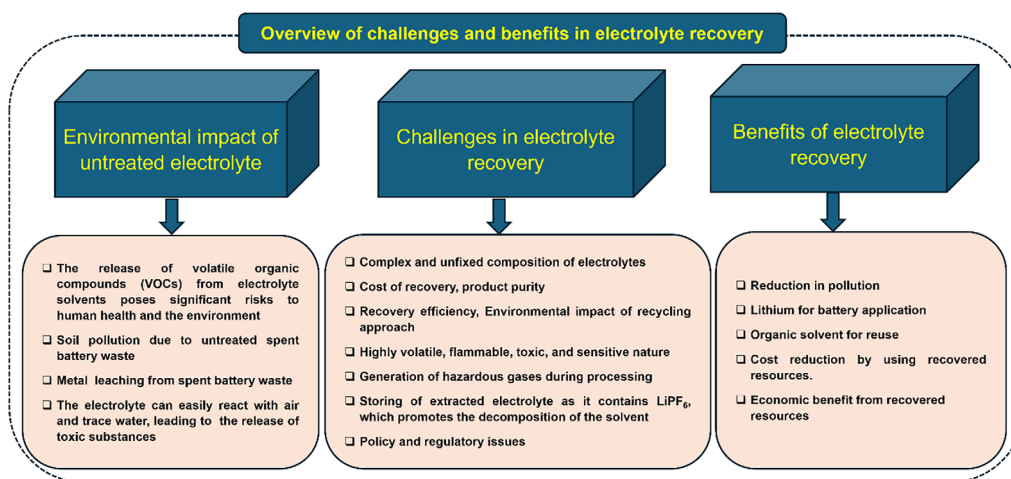#### 4.3.5. Discharge Processes Combined with Supercritical CO_2 Extraction from Aqueous Media

A more advanced extraction route utilizes supercritical CO_2 as a solvent for the selective extraction of major electrolytes from aqueous solutions.[87] Supercritical CO_2 not only enhances the efficiency of extraction but can also be less harmful to the environment, as it is nontoxic and easily separated from the extracted compounds.

Each of these approaches aims to optimize the recovery of byproducts, such as sodium fluoride (NaF) and sodium phosphate (Na_3PO_4), as well as various fuel gases, thereby making electrolyte recovery in the industry more sustainable and cost-effective.

5. Summary and Outlook

The utilization of rechargeable batteries, primarily LIBs, is steadily increasing.[15] Most likely, other emerging batteries, such as sodium-ion batteries, will also emerge on the market in the near future.[88,89] Hence, recycling spent batteries is crucial to minimizing the environmental impact of spent battery waste. Furthermore, during recycling, critical raw materials such as graphite, lithium (Li), and cobalt (Co) can also be recovered for reuse in battery manufacturing or other applications.[15]

Although recycling of cathode and anode materials from spent battery waste has gained huge attention in the past few years, but recycling electrolyte hasn't been explored at the same pace due to associated challenges such as heterogeneity in electrolyte composition, evaporation of volatile organic compounds from electrolyte, and low probability of repurposing the recovered electrolyte, which makes electrolyte recovery less attractive.[42,90] The electrolyte is a key component of a LIB, typically containing valuable $LiPF_6$ and organic solvents. Disposing of LIB waste without proper electrolyte treatment will not only create environmental problems but also result in the loss of valuable resources.[79,91] Recycling of electrolytes can provide resources such as lithium salts and organic solvents, which can be used in the lithium battery industry (**Figure 6**).[39] As Li is categorized as a critical raw material, recycling Li salts from electrolytes can make the process economically beneficial.[39,79] Furthermore, the transformation of $LiPF_6$ from electrolyte to Li_2CO_3, NaF, or Na_3PO_4, etc., could be a promising strategy to recover resources from the electrolyte.[39] Recycled organic solvents of electrolytes such as MEC, EC, and DMC have the potential to be used as fuel or solvent for a variety of material synthesis. Several scientific studies are available that utilize recovered electrolyte in battery applications. For example, Liu et al.[48] purified the spent electrolyte, and the regenerated electrolyte was used in $Li/LiCoO_2$ battery. It was reported that the efficiency of the regenerated electrolyte was very close to that of an existing commercial electrolyte with the same composition.[92] Furthermore, in another study, a vacuum distillation approach was employed for the recycling of volatile solvents (DMC and DEC) from used electrolyte, while the precipitation approach was used for the recovery of leftover lithium in nonvolatile components as Li_2CO_3.[49] Recycled volatile organic solvents were examined as the electrolyte for LIBs. It was observed that the electrolyte prepared from recycled solvent showed good cycle performance in Li/graphite battery. Recovery of electrolyte is crucial, as it can pose negative environmental impacts and safety issues due to salt decomposition and the use of inflammable, toxic organic solvents.[36] During the recovery of electrolytes, various byproducts, including wastewater, contaminated solvent, and hazardous gases, are also produced; therefore, abatement of these byproducts should also be considered. Employing green solvents during the recycling approach can

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Figure 6. An overview of environmental impact, challenges, and benefits in electrolyte recovery for LIBs.

reduce the hazardous impact on the environment. Although the supercritical extraction method for electrolyte recovery is regarded as an environmentally friendly approach, it is often associated with more costly infrastructure compared to other available methods for electrolyte recovery.[90] It's clear from the above discussion that reclaimed resources from electrolytes can be efficiently utilized in battery applications. Recovery of electrolyte will not only address the environmental issues related to spent battery waste but also provide valuable resources that can be reused in LIBs, yielding economic benefits from spent LIB waste. Therefore, further innovation is needed to develop an electrolyte recovery approach that is highly efficient, cost-effective, and has a reduced environmental impact. To establish a closed-loop battery recycling system, electrolyte recovery cannot be overlooked. It's imperative to develop an effective strategy for recycling electrolytes that reduces environmental burden and has a smaller carbon footprint during the recycling process. For the advancement of the electrolyte recovery process, making it more accurate and effective, the integration of artificial intelligence (AI) in the development of the recycling process can be beneficial, as AI can reduce trial-and-error experimentation by assisting in the systematic optimization of parameters.[93] Furthermore, integrating Internet of Things technologies into electrolyte recovery from spent batteries can help achieve consistent quality.

To bring electrolyte recovery technology from lab research to industrial-scale applications, it is essential to conduct a techno-economic analysis of the electrolyte recovery process, which will aid in the development of a business model.[94] Additionally, life cycle assessment (LCA) studies can be helpful in the development of more sustainable recycling technologies for recovering electrolytes and other valuable resources from LIBs. An LCA study can identify the key elements in the electrolyte recovery process that contribute to its environmental impact. Based on the generated database, a more effective system can be developed to reduce environmental impact during the recovery of electrolytes from battery waste.[15,95] Additionally, LCA data can provide environmental impact information on the recycling of electrolytes, which

can aid in scaling up sustainable recycling processes. For the development of sustainable battery recycling apporaches for the future, it is crucial to incorporate electrolyte recovery into the battery recycling process, enabling the creation of more environmentally friendly systems. Recovery of electrolyte materials is essential from an environmental perspective; however, more efforts focus on developing a sustainable approach for producing electrolyte materials, thereby minimizing the environmental impact of electrolyte manufacturing.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Palanivel Molaiyan: conceptualization (equal); formal analysis (equal); funding acquisition (lead); resources (equal); validation (equal); writing—original draft (equal); writing—review & editing (equal). **Hossein Rostami**: conceptualization (equal); formal analysis (equal); validation (supporting); writing—original draft (equal); writing—review & editing (equal). **Karuppasamy Karuppasamy**: data curation (equal); validation (supporting); writing—original draft (supporting); writing—review & editing (supporting). **Varsha**

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