

Designing Recyclable Electrolytes for Lithium-Ion and Next-Generation Energy Storage Devices

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The resource supply concerns raised by the rapid growth in battery production make the recycling of lithium-ion batteries (LIBs) a vital strategy for achieving a circular economy. However, current efforts mainly focus on cathode materials, while overlooking other cell components, such as the electrolyte. Commercial electrolytes, composed of costly and nonrenewable lithium salts and carbonate solvents, pose significant recycling challenges due to their high sensitivity toward hydrolysis, which complicates or even prevents recovery processes. This perspective explores the design-for-recycling concept applied to LIB electrolytes, providing insights to develop less reactive formulations compatible with simple and scalable aqueous processes. Special emphasis is

placed on the simultaneous electrolyte/electrode recovery, a strategy that could enhance the economic feasibility of electrolyte recovery while reducing the overall complexity of LIB recycling. Additionally, the recycling opportunities emerging from the early stage of development of next-generation energy storage devices are highlighted. The electrolyte recycling potential of solid-state batteries, electrochemical double-layer capacitors, and alternative battery chemistries, such as Na-ion, K-ion, or Li-S batteries, is examined. Overall, this work highlights the importance of designing electrolyte formulations with end-of-life recovery in mind to enable more recyclable energy storage devices.

1. Introduction

There is a general consensus that the electrification of the transport and power sectors through advanced energy storage devices is one of the main keys to reduce the greenhouse effect and its associated concerns. Considering Germany, in 2023, the energy sector was the largest source of greenhouse gas emissions (29%), closely followed by the transport sector (21%).^[1] These contributions can be alleviated by storing the excess of charge generated by renewable energy sources and replacing combustion engines with electric vehicles. For these purposes, lithium-ion batteries (LIBs) are the most extensively used devices due to their high-energy and long-life.^[2] However, the ongoing growth in battery production raises concerns from the resource supply perspective. While the problematics surrounding electrode raw materials are generally well known,^[3] the electrolyte components in LIBs are often overlooked. Commercial electrolytes typically consist of lithium hexafluorophosphate (LiPF₆) dissolved in a blend of linear and cyclic carbonates, such as ethylene carbonate (EC), dimethyl carbonate (DMC), or ethyl methyl carbonate (EMC).^[4] LiPF₆ is the preferred choice as the conducting lithium salt due to its high ionic conductivity, wide electrochemical stability, and controlled degradation (e.g. Al foil passivation,

solid electrolyte interphase formation).^[5] On the solvent side, EC offers a high dielectric constant, but imposes the use of cosolvents like DMC or EMC to achieve a liquid solvent media with balanced conductivity/viscosity. Additives are also included in the electrolyte formulation with the aim of fulfilling a required function (e.g. flame retardant, film formation).^[6] The manufacture of these components depends on resource-intensive and hazardous processes. Carbonate-based solvents are industrially derived from fossil feedstock-derived naphtha or natural gas. On the other hand, the production of LiPF₆ requires lithium from spodumene, phosphorus from phosphate rock, and fluorine from fluorite, involving energy-intensive reactions with H₂SO₄, Cl₂, and HF.^[7] Moreover, lithium, fluorite and phosphate rock have recently been assumed as critical raw materials by several countries.^[8] Thus, alternative solutions and innovative strategies should be considered to avoid problems in the supply distribution chain.

Likewise, as battery production intensifies, the volume of spent devices rises accordingly due to the limited service life span for EVs and small portable devices.^[9] Their improper disposal causes serious environmental and safety risks, mainly associated with the degradation of the electrolyte components. LiPF₆ can lead to soil and air contamination from highly toxic fluorine and phosphorus compounds. Also, the high flammability and volatility of the organic solvents poses additional health and environmental concerns.^[10] To address these issues, the closed-loop recycling of all LIB components is a promising approach. Recycling of LIBs can be considered as an efficient strategy to obtain the critical resources needed for their manufacture without depending on their geographical distribution. Moreover, it will considerably reduce the burden associated with their extraction and alleviate environmental concerns related to their

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disposal. Within this context, special attention should be paid to the electrolyte. Although accounting for only around 10–16 wt% of a commercial cell, electrolytes contain costly, nonrenewable materials, most notably the lithium salts.^[11] Unfortunately, the recycling of electrolyte components is nowadays greatly disregarded or not satisfactory due to certain inherent difficulties. Among them, it is worth remarking the toxic and corrosive nature of LiPF₆-based formulations, which complicates their collection, can damage equipment, and seriously affect operators' health.^[12] Recent literature reviews have summarized these problematics well while proposing novel strategies to circumvent them. All these works have agreed on the necessity for innovation regarding more recyclable electrolyte formulations.^[13–17] In this design-for-recyclability approach, electrolyte formulations would be conceptualized according to sustainable recycling methods. Unfortunately, this concept has been scarcely attempted so far and clear directions to design recyclable electrolytes are absent in literature.

Considering the aforementioned, this perspective aims to review existing LIB electrolyte recovery approaches to rationalize the challenges toward producing recyclable electrolytes. On this basis, novel strategies for the formulation of recyclable electrolytes and easier recycling methods are proposed for LIBs. Finally, the analysis is extended to next-generation storage devices, such as solid-state LIBs and post-Li-ion technologies.

2. State-of-the-Art and Current Efforts on Electrolyte Recycling

At present, spent LIBs recycling is mainly carried out through pyrometallurgical (high-temperature treatments) and hydrometallurgical (leaching of metals) processes. However, both approaches tend to focus on the recovery of cathode materials while neglecting other components of the cell, such as the electrolyte. In fact, since LiPF₆-based formulations are highly reactive, and can release hydrofluoric acid or volatile organic compounds, if not appropriately treated, most recycling workflows begin with a pretreatment step to eliminate residual electrolyte. Unfortunately, in most cases, the electrolyte is rather discarded than recovered.^[9,18] Among the few groups and companies addressing electrolyte recovery, two distinct steps can be identified: 1) enrichment, where the goal is to extract the electrolyte (and/or its decomposition byproducts) from the spent cell, and 2) resource utilization, where the extracted electrolyte components are purified or transformed for reuse.

The enrichment step's main challenge is the extreme sensitivity of LiPF₆ toward moisture, as it rapidly releases HF and POF₃ when in contact with water traces. As a result, conventional aqueous or thermal methods often degrade LiPF₆, complicating any effort to isolate the intact electrolyte. Additionally, this limits the range of electrolyte extraction techniques and increases the price and complexity of the overall recycling process. Thermal (freezing, evaporation), mechanical (centrifugation, positive/negative pressure), electrochemical (electrodialysis), and chemical

(solvent extraction, supercritical CO₂ extraction) methods have been explored to surpass this challenge.^[19] Nevertheless, most of these techniques still decompose the LiPF₆ salt, ignore the recovery of the solvents, and involve the use of complex instrumentation or controlled environments. Among them, solvent extraction, sometimes combined with supercritical CO₂ extraction, presents the best capabilities so far since allowing the recovery of the electrolyte as a whole. Moreover, it also facilitates the extraction of the electrolyte fraction trapped in other components of the cell, such as the separator or the electrodes.^[20] Still, this process requires complex instrumentation and strictly anhydrous organic solvents, limiting its large-scale implementation.

During the resource utilization step, the mixture of recovered electrolyte components (or their degradation products) is placed back in the value chain. Generally, compound recovery is first pursued with the aim of isolating the individual components of the electrolyte through methods such as liquid–liquid phase separation, distillation, or chemical precipitation. As follows, the reclaimed compounds are revalorized. Ideally, regeneration and direct reutilization will be preferred since reducing costs and lowering the associated ecological footprint.^[21] In practice, the high reactivity of LiPF₆, especially after its partial hydrolysis during the enrichment step, renders the full regeneration of battery-grade LiPF₆ extremely difficult. Accordingly, most processes deliberately decompose LiPF₆ into more stable, high-value byproducts, such as Li₂CO₃.^[22,23] On the other hand, solvent mixtures from the electrolyte (e.g. EC, DMC, EMC) are difficult to separate and a lot of energy is required for their purification. Due to these challenges and their relatively low market value, carbonate-based solvents are generally burnt or simply discarded.^[1,2]

3. Design for Recycling of Electrolytes

Although great efforts have been made, it is clear that electrolyte recycling remains a stumbling block in achieving LIBs closed-loop recycling due to the high reactivity of LiPF₆-based formulations. Ideally, direct recovery and reutilization of the electrolyte components in near-battery-grade form will be preferred. Thus, minimizing electrolyte decomposition during the enrichment step is crucial. This would enable the use of simple and scalable solvent extraction techniques, that do not necessarily rely on expensive anhydrous chemicals (i.e. water, acetonitrile) to recover the electrolyte components and their side products as a whole. Minimizing electrolyte components reactivity would also support the implementation of efficient separation and regeneration techniques, thereby enabling the reclamation of battery-grade electrolyte components. On the other hand, nowadays electrolyte recovery is considered as a separate step from the whole recycling process. Thus, adapting solvent extraction to simultaneously tackle the recovery of other elements in the cell, such as the active materials of the electrodes, would also bring several ecological and economic benefits. With this in mind, tailored electrolyte formulations compatible with competitive solvent extraction approaches and simple separation procedures, allowing the recovery of the electrolyte fraction and other components of

the cell, should be strongly considered. The following sections are devoted to propose alternative recyclable formulations compatible with the aforementioned sustainable recycling approaches.

3.1. Formulation of Water-Stable Liquid Organic Electrolytes for LIBs

The use of water as an extraction solvent is a promising alternative to conventional organic extractants due to enabling the use of well-known methodologies such as liquid–liquid phase separation or reduced vapor distillation. Thus, the formulation of water-resistant electrolytes cannot only greatly simplify the overall recycling process but also reduce health and environmental hazards. Obviously, water processes are not compatible with benchmark LiPF_6 -based formulations, mainly due to the high reactivity of F. The utilization of alternative elements in electrolyte salt compositions could prove beneficial from a recycling perspective. Different fluorine-free lithium salts composed of perchlorates, phosphates, borates, and heterocyclic anions have been proposed.^[24] However, the complete replacement of fluorine is challenging due to its intrinsic benefits toward cell operation.^[25] On this regard, the well-established imide-based salts lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) and lithium bis(fluorosulfonyl)imide (LiFSI) present promising opportunities. Besides their well-known positive features for cell operation, these salts demonstrate considerably lower (LiFSI) or even negligible (LiTFSI) sensitivity toward hydrolysis when compared to LiPF_6 .^[26] Among them, LiFSI has recently attracted attention since recent advances on its large-scale production lead to a gradual increase of its portion in commercial electrolyte formulations.^[27] In contrast, LiTFSI remains more expensive and its large-scale adoption will require further cost reductions or the successful development of efficient recycling strategies.^[28] Surprisingly, although imide-based salts, and specially LiFSI, are considered

next-generation salts, examinations of its recyclability are glaringly absent in literature. Unfortunately, imide-based salts are unable to protect the Al current collector from anodic dissolution at high potentials. This challenge can be solved through the incorporation of multifunctional film-forming additives, such as lithium bis(oxalato)borate (LiBOB) to improve cell performance without compromising recyclability. Despite presenting some hydrolytic activity, decomposition products of LiBOB are much less toxic and corrosive when compared to these of LiPF_6 . Moreover, LiBOB can scavenge small amounts of HF, making it an ideal partner for salts that are prone to release small amounts of HF when exposed to water, such as LiFSI.^[29] Unfortunately, there are a lack of detailed studies dealing with the hydrolysis extent and mechanisms of lithium salts under industrial conditions (i.e. large amounts of water, wide temperature range, different pH evolution).^[30] The influences of pH and temperature over these electrolytes and the use of alternative cheap solvents (e.g. acetonitrile) could lead to fast improvements in their recyclability. Advantages and challenges toward the implementation and recycling of various lithium salts are summarized in **Table 1**.

Regarding the electrolyte solvents, carbonate blends are compatible with water-extraction strategies. Moreover, recent studies have demonstrated that the release of volatile compounds can be mitigated by immersion of spent batteries in water.^[31] After water-assisted electrolyte extraction from the spent cell, liquid–liquid phase separation methods can be applied to recover the lithium salt in the water phase while isolating the electrolyte solvents in the organic phase. For this, it is crucial to identify suitable organic extractants able to form immiscible phases while exhibiting a lower boiling point than the electrolyte solvents. Extraction of the electrolyte components from the water and organic phase can be following pursued through simple methods, such as reduced pressure distillation. The presence of these extractants in the organic solvents after the distillation should be

Table 1. Summary of relevant properties for the implementation and recycling of various lithium salts.

Type	Li salt	Challenges	Advantages
Benchmark	LiPF_6	Strong hydrolysis, HF release Serious environmental concerns Noncompatible with acid-sensitive solvents/additives	Established production and implementation Film-formation abilities Good associated conductivity
Fluorine-free	LiBOB	Low associated conductivity Significant release of gases Low solubility in carbonates	Simple production route, low price Moderate hydrolysis, benign decomposition products Film-formation abilities
	LiClO_4	Low safety, explosion risks Highly corrosive	Simple production route, low price Stable in air and water
Imides	LiTFSI	Complex and costly production Unable to passivate Al	High resistance toward hydrolysis Good associated conductivity Compatible with several chemistries
	LiFSI	Unable to passivate Al Hydrolyzable at high temperature or basic/acid conditions	Good associated conductivity Established production accompanied with price reduction
Others	LiBF_4	Low-associated conductivity Middle hydrolysis	Middle film-formation abilities High thermal stability
	$\text{LiDFOB}^{\text{a)}$	Large-scale production nondemonstrated Middle hydrolysis Low-associated conductivity	Film-formation abilities Higher solubility than LiBOB in carbonate-based solvents

^{a)}Lithium difluoro(oxalato)borate.

minimized to ensure the proper reutilization of both components (or mixture of components). However, difficulties in separating and purifying carbonate solvents, along with the low economical profitability of their recovery, could compromise their recycling. Therefore, replacing benchmark carbonates with bioderived alternatives relying on noncritical materials should be prioritized. The development and application of these sustainable solvents, such as gamma-valerolactone (GVL), has made considerable advances in the recent years.^[32–34] However, the range of bio-based alternatives is still limited and challenges regarding performance need to be overcome. Finding novel bio-derived solvents or developing novel strategies for the production of conventional carbonates from renewable sources could critically reduce the overall carbon footprint of LIBs. Moreover, the selection of the solvents could be guided by their potential repurpose in less-demanding applications. For example, GVL has demonstrated potential to replace the toxic N-methyl pyrrolidone (NMP) in electrode manufacturing processes.^[35]

Summing up, identifying novel water-resistant salts and understanding their hydrolytic decomposition routes are essential for developing water-stable electrolytes compatible with simple and sustainable recycling approaches. This would enable the use of water for the extraction of the electrolyte from spent batteries. Following, liquid–liquid-phase separation methods can be applied using suitable extractants to isolate the solvent in the organic phase and the lithium salt in the water phase. Techniques such as reduced vapor pressure distillation can be employed to separate the electrolyte solvent from the extractants and the lithium salt from the water phase. Recovered lithium salts could be directly reutilized, while organic solvents can be repurposed for less-demanding applications in case that the presence of impurities hinders their direct reutilization in LIB applications. The suitability of this approach was recently demonstrated by our group through the successful formulation, recycling, and reutilization of a tailor-made electrolyte consisting of LiTFSI and GVL (Figure 1).^[36]

3.2. Simultaneous Electrode/Electrolyte Recovery

The electrolyte recovery is typically addressed as a separate step by the few companies and groups addressing this topic. This disconnection not only undermines the economic feasibility for the recovery of the electrolyte components but also increases the complexity of the overall recycling process. Integrating electrolyte recovery with the extraction of other elements in the cell may represent a significant step forward to achieve the closed-loop recycling of LIBs. Even if promising, just a few studies have explored this concept.^[37,38] The work of He et al. demonstrated the feasibility to adapt the electrolyte extraction method to simultaneously recover the electrode active materials.^[38] Nevertheless, LiPF₆ decomposition was not completely avoided, leading to the partial formation of Li₂CO₃. This underscores the high reactivity of conventional electrolyte components, hindering their recovery.

However, the current market demand on replacing conventional organic solvent-based to water-based casting processes presents a valuable opportunity to merge electrolyte and electrode materials recovery through sustainable approaches. Water-based electrode manufacturing processes present enhanced sustainability over conventional methods, which rely on the use of toxic NMP as solvent and highly fluorinated polyvinylidene fluoride (PVDF) as binder.^[39] Additionally, their implementation would reduce the considerable associated costs for NMP drying. Nevertheless, compatibility with aqueous processing is still limited for some cathode chemistries, which are prone to ion leaching in water. However, this is not the case for LiFePO₄ (LFP), which exhibits relatively good water stability. Its implementation in EVs is steadily increasing due to its lower cost and higher safety, making its recycling appealing.^[40] To this end, a new generation of hydrolyzable binders has been developed, paving the way for the simple recovery of electrode active materials upon water addition. Furthermore, mechanical separation methods (e.g. flotation, centrifugation) enable the separation of positive

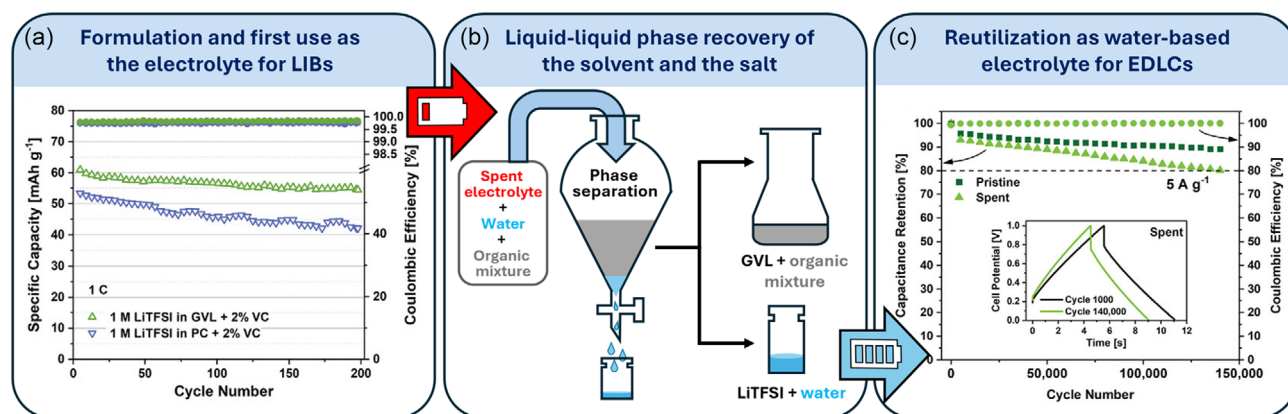


Figure 1. a) long-term galvanostatic cycling of 1 M LiTFSI in GVL electrolyte for LIBs compared with a standard formulation, b) the liquid–liquid phase process for the recovery of the solvent in the organic phase and the salt in the water phase, and c) the reutilization of the recovered LiTFSI in water solution for EDLCs. Modified with permission.^[37] Copyright 2025, Wiley.

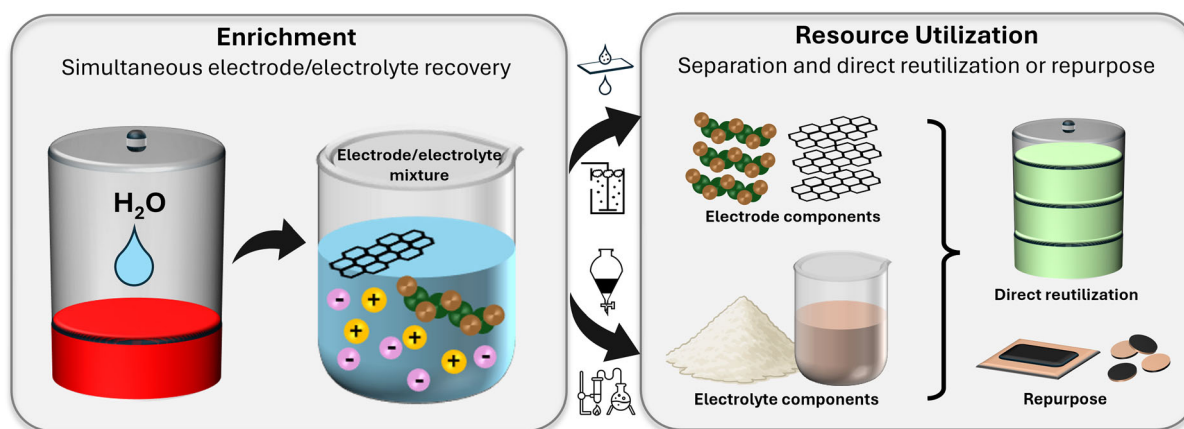


Figure 2. Diagram illustrating the recycling opportunities of developing hydrolysis-resistant electrolytes.

and negative electrode materials without compromising their chemical structure.^[41] From this perspective, the formulation of novel water-tolerant electrolytes should be also strongly considered due to allowing the simultaneous recovery of both the electrolyte components and active materials from the electrodes (Figure 2).

Despite these opportunities, several challenges remain to achieve truly integrated recycling strategies that are applicable under real-world conditions, such as developing water-stable electrolytes and electrodes with competitive performance. Additionally, the possible interferences caused by the dissolution of water-soluble binders, the presence of conductive additives, or the leaching of metals from the current collectors require detailed investigations and the establishment of mature separation processes.^[42] Another critical aspect lies in the enhanced sensitivity of aged cathode materials to water, even for those generally considered water stable, such as LFP. After prolonged cycling, the formation of small cracks can expose fresh active surfaces to water,^[43] promoting ion leaching and the formation of insulating surface layers. These structural and chemical changes can significantly hinder the electrochemical performance of recovered materials. Therefore, water-assisted extraction processes must be carefully designed to minimize the risk of further active material degradation. Fortunately, there are well-known regeneration techniques that are particularly compatible with water-based processing that could be effectively coupled with electrolyte recovery schemes. For example, direct regeneration of the LFP structure can be achieved in water through the addition of mild reducing agents and inexpensive Li salts.^[44] However, to properly guide regeneration strategies and reduce active material degradation during the recycling process, it is critical to better understand the aging mechanisms and surface–interface formation processes of cathode materials in alternative electrolyte formulations.^[45]

3.3. Recycling of Solid-State Electrolytes

Solid-state electrolytes (SSEs) have been extensively studied in the past few decades since providing enhanced thermal stability,

reduced flammability, and limiting the shortcomings associated with dendrite growth in energy-dense lithium metal anodes. Moreover, the paradigm shift in LIB design driven by the potential implementation of SSEs presents a valuable opportunity to improve their recyclability at this early stage of development.^[46,47] Among SSEs, both inorganic solid electrolytes (ISEs) and solid polymer electrolytes (SPEs) present inherent advantages and challenges.

Regardless of their type (e.g. oxides, sulfides, halides), ISEs do not require the incorporation of a lithium salt since Li ions are already integrated within their structure. This potentially avoids the separation step of the electrolyte components and facilitates its direct reutilization. Among them, garnet-type oxide electrolytes, such as lithium lanthanum zirconium oxide (LZO), present promising features. Their good stability under ambient conditions and relatively higher resistance toward hydrolysis enables their recovery under simple conditions. Unfortunately, their complete mechanical recovery would not be possible since a certain amount of these oxides needs to be included also in the electrode formulation to provide sufficient lithium transport. Despite this, hydrothermal recycling methods currently in use for cathodes can be adopted with relative simplicity for these types of electrolytes due to their chemical similarities.^[47] Additionally, several works have reported the feasibility of direct reutilization of garnet-type electrolytes through simple relithiation and thermal strategies.^[48–50] Among these, simultaneous hydrothermal regeneration of both electrodes and garnet-type electrolytes present several opportunities due to its environmental friendliness and reduced cost.^[51,52] On the other hand, the recycling of non-water-resistant sulfide-type ISEs has also been demonstrated to be suitable and practical. Their separation from the rest of the components of the cell can be achieved by dissolution with the use of cheap and abundant organic solvents (e.g. ethanol, acetonitrile). Following, the sulfide-based SSE chemical structure can be recovered through reprecipitation and recrystallization.^[53–55]

Regarding SPEs, their recovery can be more challenging, since a lithium salt needs to be integrated within the polymer host to enable Li⁺ transport. Depending on the selected salt, the recycling process can suffer from the same challenges of

conventional organic electrolytes. Additionally, the separation of the lithium salt from the polymer host can be likewise challenging. Fortunately, aqueous-resistant salts, such as LiTFSI, are employed in most of the cases, which enable the integration of water-extraction technologies.^[56] Furthermore, the great tunability of polymers permits the design of molecular structures adapted to favor simple separation processes. For example, recent studies have successfully applied the design-for-recyclability strategy by developing thermally or chemically depolymerizable polymer hosts. These approaches successfully enabled the recovery of LiTFSI in acceptable yields and proved its reutilization.^[57,58]

3.4. Perspectives for Post-Lithium-Ion Batteries

To advance in sustainability, great efforts are currently made to develop alternative batteries to LIBs that use less critical raw materials, such as sodium-ion (SIBs) or potassium-ion (KIBs) batteries. However, similar production routes are followed for the manufacture of their benchmark salts (i.e. NaPF₆, KPF₆) and solvents (i.e. EC, DMC, EMC).^[59] The recycling of the electrolytes for these devices will be eventually desired, facing the same challenges of LIB formulations. So far, the recycling of alternative energy storage electrolytes has been scarcely attempted. Nevertheless, the lower sensitivity toward hydrolysis of Na-based and K-based salts represents a great opportunity. In contrast to LiFSI, their Na and K analogs present considerably reduced hydrolysis susceptibility, serving as promising components for the formulation of water-recyclable electrolytes.^[30] Besides, the aqueous recycling processes are also relevant to lithium–sulfur (Li–S) batteries. Benchmark Li–S electrolytes already employ a water-stable imide-based salt (LiTFSI),^[60] making these devices ideal candidates for water-based recycling approaches. However, special attention needs to be paid to the dissolution of sulfur species in the electrolyte, which could greatly undermine the separation and purification processes.

The development of sustainable recycling processes is interesting as well for electric double layer capacitors (EDLCs),

specifically for those employing water-in-salt electrolytes, in which a prohibitive amount of imide salt (LiTFSI) is used.^[28] In this regard, the absence of organic solvents could significantly simplify the recovery process. Additionally, a previous study already demonstrated the feasibility for the recovery of LiTFSI from a concentrated electrolyte formulation.^[61] However, further studies are required to understand the formation of decomposition products during cell operation that may affect the recycling procedure. The design of simple and scalable processes could also promote the implementation of ionic liquids (ILs) for EDLCs. The high cost associated with the production of ILs could be alleviated if a proper recycling procedure is demonstrated. Among the wide range of ILs, those containing more hydrophobic anions, such as TFSI[−], could simultaneously enhance the voltage window of the device while being compatible with sustainable recycling approaches (Table 2).^[62]

4. Discussion

Current electrolyte recycling techniques represents a bottleneck in achieving the complete and closed-loop recycling of LIBs due to the high reactivity of commercial LiPF₆-based electrolytes. To address this, the formulation of novel electrolyte configurations with reduced reactivity, particularly those presenting reduced hydrolytic activity, can facilitate the implementation of easier, scalable, and sustainable recycling methods. Fluorine-free and hydrolysis-resistant imide-based salts should be prioritized, because of offering enhanced sustainability and recyclability. On the other hand, the use of bioderived solvents can improve the overall sustainability of the devices. Moreover, solvent selection can be guided by considering their potential repurposing in less demanding applications, such as electrode manufacturing processes. The implementation of such water-resistant formulations would enable the use of simple and scalable enrichment and separation processes, such as water-extraction strategies followed by liquid–liquid phase separation and reduced pressure distillation. Importantly, the formulation of water-stable

Table 2. Challenges and opportunities for the recycling of next-generation energy storage devices.

Device	Electrolyte	Challenges	Opportunities
SSEs (LIBs)	Garnet-type	SSE also included in electrode formulations	Stable under air and moisture Compatible with cathode hydrometallurgical methods
	Sulfide-type	Release H ₂ S in contact with H ₂ O	Compatible with dissolution and recrystallization processes
	Polymeric	Separation of the Li salt and the polymeric host	Nonhydrolyzable salts generally used Easy tunability of the polymeric host to separate electrolyte components
EDLCs	Water-in-salt	No studies on the influence of salt concentration over recycling	No organic solvents in the formulation Compatible with aqueous-processing
	Ionic liquids	Lack of studies on the hydrolytic activity of ILs	ILs containing hydrophobic anions may be compatible with aqueous processes
SIBs / KIBs	Liquid organic electrolytes	No studies on the influence of the cation over the recycling process	Na and K salts more stable toward hydrolysis than their Li counterparts
Li-S batteries	Liquid organic electrolytes	Dissolution of sulfur species in the electrolyte	Compatible with aqueous processing

electrolytes also opens the door to the simultaneous recovery of electrolyte and electrode materials. In such strategy, water serves both as the extraction solvent for electrolyte recovery and as the exfoliation agent to delaminate active electrode materials from their current collectors. This synergy could reduce process complexity, improve electrolyte recycling economic feasibility, and align with green manufacturing principles. For next-generation energy storage devices, their early stage of development presents a valuable opportunity to incorporate design-for-recycling principles. Among them, SSEs present inherent features that facilitate their recovery. While garnet-type SSEs are compatible with existing hydrometallurgical processes used for cathodes, sulfide-type SSEs can be easily recovered using inexpensive organic solvents and recrystallized with minimal degradation. On the other hand, the great tunability of SPEs allows for the development of polymeric matrices with tailored properties to enhance their recyclability, especially by enabling controlled decomposition for the separation of electrolyte components. For SIBs and KIBs, the lower sensitivity toward hydrolysis of their salts (NaFSI, KFSI) further enhances the prospects for water-based recycling strategies. Thus, these systems represent an advantageous platform for implementing early-stage design-for-recycling principles. These principles can also be extended to EDLCs and Li-S batteries, which generally rely on water-stable salts that can be easily recovered through the aforementioned processes.

5. Conclusions

This perspective provides pathways to overcome the electrolyte recycling bottleneck by rethinking conventional formulations as a gateway to simpler, safer, and more scalable recovery methods. To achieve this, guidelines are given for the design of tailored formulations compatible with water-based methodologies through the selection of stable salts and repurposable solvents. Special emphasis is placed on the use of water as both an extraction medium and an exfoliation agent, paving the way for simultaneous electrode/electrolyte recovery. Additionally, the unique opportunities to integrate recyclability from the outset of emerging energy storage devices are highlighted for solid-state batteries, SIBs, KIBs, Li-S batteries, and EDLCs. Overall, these design-for-recyclability approaches for electrolytes could significantly reduce reliance on critical materials while advancing the goal of closed-loop recycling for energy storage devices.

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Conflict of Interest

The author declares no conflict of interest.

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