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Toward closed-loop hydrometallurgy: a critical review of wastewater reuse strategies for end-of-life $LiFePO_4$ battery recycling

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The rapid adoption of $LiFePO_4$ (LFP) batteries, driven by their safety and cost advantages, necessitates the development of sustainable recycling technologies tailored to their low-value composition. While hydrometallurgical methods enable efficient lithium recovery, they typically generate large volumes of wastewater, undermining both environmental and economic viability. This review critically examines recent advances in closed-loop hydrometallurgical recycling of end-of-life LFP batteries, with a particular focus on wastewater reuse strategies. A comprehensive classification of lixiviants, including inorganic acids, salt-based compounds, and alkaline and organic compound, is presented, highlighting wastewater reuse. Quantitative techno-economic simulations reveal that full wastewater reuse, starting from the second cycle, significantly reduces operational costs and enables positive net profit within four to five reuse cycles in regions with favorable energy and labor conditions. Despite these advancements, challenges such as impurity build-up, reagent stability, and limited long-term cycle testing remain. This review outlines current limitations and proposes future directions for scaling up closed-loop systems in alignment with the principles of green chemistry and circular hydrometallurgy.

Green foundation

1. This review highlights recent advances in closed-loop hydrometallurgical recycling of $LiFePO_4$ (LFP) batteries, focusing on wastewater reuse strategies. It presents a classification of lixiviants and demonstrates how reusing process water from the second cycle onward can significantly reduce chemical use, waste generation, and overall costs.
2. As LFP batteries continue to gain market dominance, current recycling systems, which are designed for cobalt- and nickel-rich batteries, are no longer sufficient. This review addresses the urgent need for sustainable solutions tailored to low-value battery chemistries, such as LFP, making it relevant to industry, policymakers, and researchers alike.
3. The future lies in fully closed-loop processes that integrate wastewater reuse. This review outlines key challenges, such as impurity buildup and reagent stability, and provides guidance for developing scalable, green, and economically viable recycling systems that align with the principles of circular hydrometallurgy.

1. Introduction

The global lithium-ion battery (LiB) industry is undergoing a notable transformation, marked by a gradual yet steady shift from nickel- and cobalt-rich chemistries, such as lithium nickel manganese cobalt oxide ($LiNiMnCoO_2$; NMC), toward lithium iron phosphate ($LiFePO_4$; LFP) batteries.[1,2] This shift is largely driven by the favorable cost-performance profile of LFP, which includes superior thermal stability, longer cycle life, high safety under abusive conditions, and the absence of expensive and geopolitically sensitive metals such as cobalt and nickel.[3] According to the IEA report,[4] LFP batteries accounted for over 40% of the global electric vehicle (EV) battery capacity in 2023, maintaining a strong competitive edge in terms of cost-effectiveness and thermal stability. By 2024, their market share increased to nearly 50% globally, driven largely by widespread adoption in China, where LFP batteries were used in approximately 75% of all EVs sold.[4] Although official data for 2025 are not yet available, current trends suggest that LFP will continue to sustain a dominant position, with its global market share expected to remain within the 40–50% range.[4] Despite this rapid growth, current battery recycling technologies and regulatory frameworks have

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largely been developed around NMC-type chemistries, primarily due to the high economic value of cobalt and nickel.[3,5,6] As a result, existing industrial recycling infrastructures, particularly hydrometallurgical processes, are typically optimized for the recovery of high-value metals such as cobalt and nickel,[7–10] resulting in a significant technological and economic gap in the recycling of end-of-life (EoL) LFP batteries. Moreover, since LFP contains lithium and iron, the conventional value-recovery logic that justifies recycling is substantially weakened.[11,12] In light of these challenges, the development of sustainable and efficient recycling solutions specifically tailored for LFP batteries has become increasingly urgent. Nonetheless, existing recycling approaches for LFP remain largely at the laboratory scale, with significant hurdles to industrial scalability.[11] Among the various technologies for EoL LFP battery recycling, including pyrometallurgical, hydrometallurgical, and direct recycling methods,[13–16] this review focuses on hydrometallurgy due to its compatibility with low-temperature processing and potential for the selective and efficient recovery of lithium.

One of the most prominent barriers to the implementation of large-scale hydrometallurgical recycling for LFP batteries is its limited economic viability.[11,17,18] Unlike NMC-type cathodes, which contain high-value metals such as cobalt and nickel, LFP is composed primarily of lithium, iron, and phosphate elements with relatively low market prices (Fig. 1). In particular, iron and phosphate have minimal resale value in secondary markets, while lithium, though previously considered a strategic resource, has experienced significant price volatility in recent years.[19] For instance, the spot price of lithium dropped from about \$80 000 per metric ton in late 2022 to about \$8300 in June 2025,[20] substantially undermining the economic rationale for lithium recovery alone. Several techno-economic assessments reinforce this challenge. According to the analysis, the net profit from conventional hydrometallurgical recycling of EoL LFP cathode materials varies significantly by region, ranging from approximately −\$11.8 (UK) to −\$4.9 (China) per kWh,[18] indicating an overall negative profitability. In comparison, the recycling of EoL NMC cathodes (*e.g.*, NMC622) demonstrates positive economic returns, with net profits ranging from approximately \$2.91 to \$9.83 per kWh,[18] primarily attributed to the high market value of recovered cobalt and nickel (Fig. 1). These economic limitations are also reflected in the process design. For instance, solvent extraction, a liquid–liquid separation technique, is widely employed in hydrometallurgical processes to selectively transfer target metal ions from an aqueous leachate into an immiscible organic phase containing a specific extractant. This method is particularly effective for achieving high extraction efficiency and product purity, especially when treating a complex leachate containing multiple metals. In the case of NMC battery recycling, solvent extraction plays a critical role in isolating and purifying high-value metals such as Ni and Co from solution (Fig. 1a). By contrast, the hydrometallurgical recycling of LFP batteries generally involves a much simpler

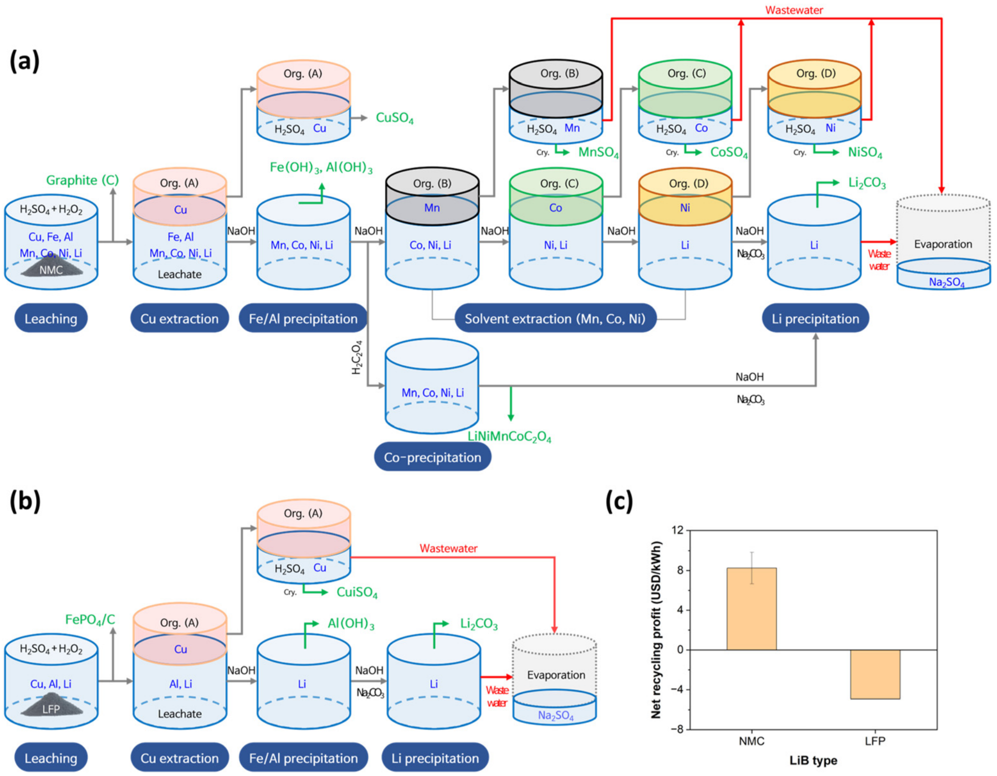


Fig. 1 Hydrometallurgical recycling flowsheet examples and economic comparison for different LiB chemistries: (a) NMC battery recycling process; (b) LFP battery recycling process; (c) net recycling profit per kWh for NMC (average across types) and LFP batteries using data reported by Lander *et al.*[18]

process, since lithium is considered the only economically valuable element for recovery (Fig. 1b). As a result, solvent extraction is rarely employed in LFP recycling workflows. When used, it is typically limited to removing non-valuable impurities such as aluminum or copper from the leachate. However, even these can often be precipitated as hydroxides by simple pH adjustment, thereby reducing the need for organic solvent consumption. Nonetheless, the recycling of LFP batteries cannot be dismissed purely on economic grounds. From a policy and environmental perspective, LFP recycling is increasingly seen as essential to achieving resource circularity, reducing landfill burden, and minimizing the environmental footprint of LiB waste.[21,22] Furthermore, upcoming battery regulations in regions such as the European Union (EU) and China are expected to mandate the recovery of lithium and other critical raw materials regardless of their market value.[23]

In addition to limited economic returns, one of the most critical drawbacks of hydrometallurgical processes is the generation of large volumes of wastewater containing dissolved salts and unreacted reagents.[24–26] Unlike pyrometallurgical routes, which primarily produce concentrated solid residues, aqueous hydrometallurgy inherently depends on extensive liquid phase operations such as leaching, washing, neutralization, and separation, each of which contributes to the accumulation of complex liquid waste streams. The composition of wastewater generated during LFP hydrometallurgy is strongly influenced by the type of lixiviant employed and by the alkaline reagents, such as NaOH, Na_2CO_3, or Na_3PO_4, used during the lithium recovery step. For example, processes utilizing sodium salt-based lixiviants such as sodium bisulfate ($NaHSO_4$),[27] sodium persulfate ($Na_2S_2O_8$),[28,29] or sodium dihydrogen phosphate (NaH_2PO_4)[22,30] lead to wastewater enriched with Na^+ and anionic species including PO_4^{3-} and SO_4^{2-}. Furthermore, neutralization reactions often generate large amounts of sulfate-rich brines (*e.g.*, Na_2SO_4), which typically require additional treatment.[25]

Conventional hydrometallurgical research on the recycling of EoL LFP batteries has focused on four key areas: (1) selective

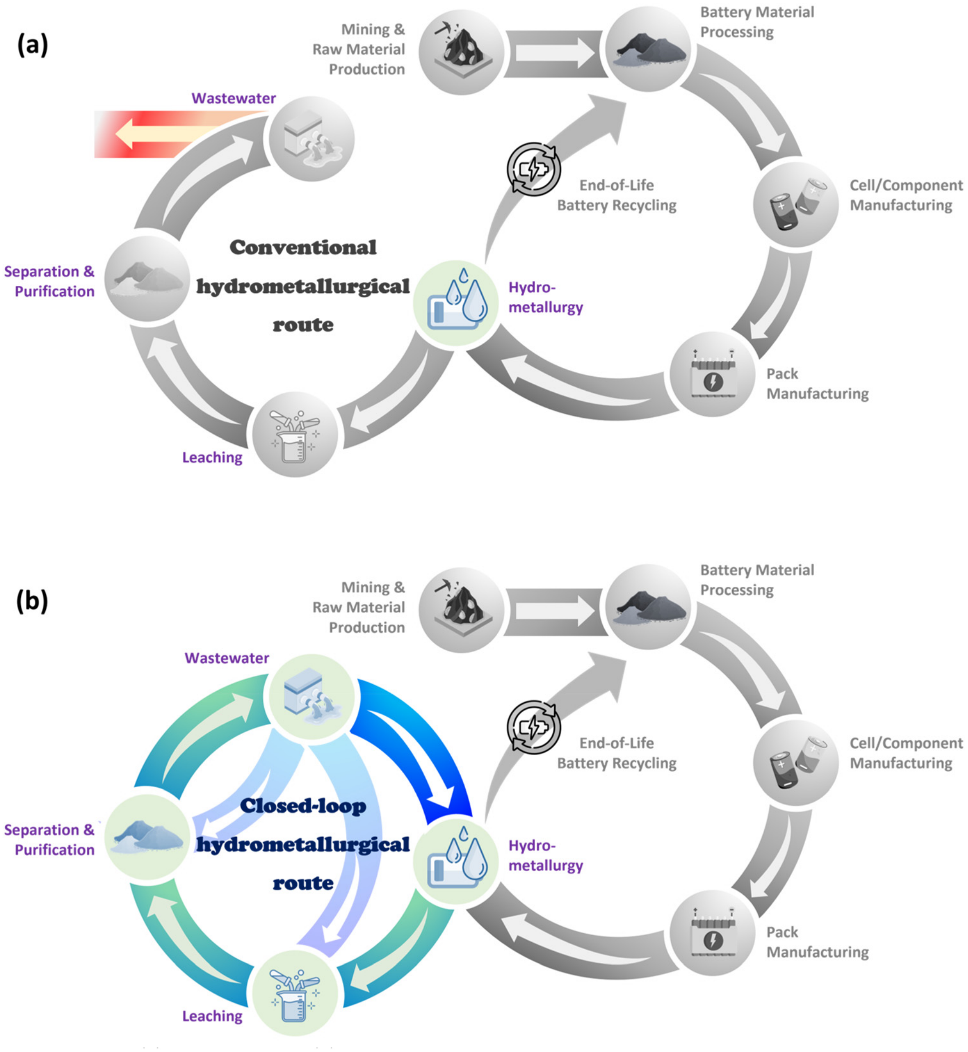


Fig. 2 Conceptual comparison between (a) conventional and (b) closed-loop hydrometallurgical recycling routes in a circular LiB system.

lithium leaching using either inorganic acids or environmentally benign solvents; (2) recovery of high-purity metal compounds such as Li_2CO_3 and $FePO_4$; (3) re-synthesis of LFP (S-LFP) cathodes utilizing as-recovered metal compounds; and (4) electrochemical performance evaluation of S-LFP. Collectively, these research efforts have demonstrated the potential of hydrometallurgical recycling to contribute to a closed-loop material supply chain for LFP-based lithium-ion batteries (Fig. 2). However, despite these advances in material recovery and reuse, the hydrometallurgical processes themselves are often not circular in a broader environmental or process-integrated sense (Fig. 2a). In most cases, lithium leaching and separation are accompanied by the generation of substantial volumes of wastewater containing residual reagents and dissolved salts. This not only imposes a considerable environmental burden but also undermines the overall economic and ecological viability of the process. As a result, although the reuse of recovered lithium and iron compounds supports material circularity, conventional LFP hydrometallurgy fails to achieve true closed-loop operation at the system level. Addressing this shortcoming requires expanding the scope of research from product-focused recovery to include wastewater reuse and process-level circularity, which can enhance both sustainability and cost-effectiveness (Fig. 2b).

In summary, from both environmental and operational perspectives, wastewater treatment represents a significant cost and bottleneck in LFP recycling workflows. Therefore, shifting the paradigm from "treatment and disposal" to "reuse and recirculation" offers a compelling opportunity to enhance sustainability. These benefits not only improve the environmental profile of hydrometallurgical processes but also strengthen resource efficiency by minimizing wastewater treatment demand and reducing the need for additional chemical reagents. This shift in focus aligns with the "Close Water Loops" principle outlined in the Twelve Principles of Circular Hydrometallurgy reported by Binnemans and Jones,[31] which emphasizes the reuse of water and process liquors within hydrometallurgical circuits to minimize freshwater consumption and wastewater disposal. As emphasized in this principle, closing the water loop is not only a technical challenge but also a strategic imperative for achieving sustainable and economically viable recycling systems,[31] especially for low-value materials such as those found in LFP batteries. Hence, this review seeks to address this gap by systematically compiling and evaluating closed-loop hydrometallurgical approaches that integrate wastewater recycling into the process flow, thereby transforming waste streams into functional resources. Specifically, this review categorizes existing research into two main clusters: a conventional hydrometallurgical route and a closed-loop hydrometallurgical route *via* wastewater reuse. The former section (section 2) outlines representative case studies reported so far on hydrometallurgical LFP recycling. The latter section (section 3) presents studies in which wastewater streams are directly reused. Finally, this review provides an in-depth insight into the research needs for realizing "fully" closed-loop hydrometallurgical recycling of LFP batteries.

2. Conventional hydrometallurgical approaches for recycling of EoL LFP batteries

Before delving into the main content of this section, EoL LFP batteries typically undergo a series of pre-treatment steps, such as discharging, shredding, and material separation, to obtain black powders (also known as black mass) that contain the valuable metals. The pre-treatment procedures for LFP batteries have been described in detail in our group's recent publication.[29] These operations are essential for enhancing leaching efficiency and minimizing contamination in downstream processes. While this review focuses on the post-treatment stage, *i.e.*, hydrometallurgy, the role of pre-treatment is duly acknowledged as a critical enabler of effective and safe recycling.

Hydrometallurgical processes have been extensively applied to the recycling of EoL batteries, due to their effectiveness in recovering target metals, as shown in Table 1. These processes typically involve a sequence of unit operations, including leaching and separation (generally precipitation for lithium recovery). So far, as previously discussed, hydrometallurgical recycling of LFP has primarily focused on four key areas to validate its suitability for reuse in battery manufacturing (Fig. 1a).

A primary focus of conventional hydrometallurgy for recycling EoL LFP batteries lies in achieving high metal leaching efficiency (LE_i; i = element), often exceeding 95%, through the use of various lixiviants (Table 1). The leaching behavior is governed by the thermodynamic stability of LFP, which is determined by pH and redox potential (E_h) as shown in the Pourbaix diagram of the Li–Fe–P–H_2O system (Fig. S1).[29,44] For instance, in acidic environments without the presence of oxidants, LFP undergoes simultaneous dissolution of both Li^+ and Fe^{2+}, leading to the co-existence of these ions in the leachate. This process occurs through proton-assisted breakdown of the LFP's olivine structure, which results in poor selectivity due to the undesired co-leaching of iron. A representative reaction using sulfuric acid (H_2SO_4) can be described as follows:[45]

$$2LiFePO_4(s) + 3H_2SO_4(aq) \rightarrow Li_2SO_4(aq) + 2FeSO_4(s) + 2H_3PO_4(aq). \quad (1)$$

More than 90% $LE_{Li\&Fe}$ was reported using H_2SO_4 (*e.g.*, 2.5 M H_2SO_4 at 60 °C for 4 h, solid–liquid (S/L) = 10 mL g^{-1}), as demonstrated by Zheng *et al.*[33] and Song *et al.*,[35] while hydrochloric acid (HCl)-based systems also achieved similar results when combined with hydrogen peroxide (H_2O_2) as an additive.[37,38] Organic acids such as methanesulfonic acid (CH_4O_3S) have been successfully applied under mild conditions, achieving over 94% lithium and iron leaching.[42] In these systems, when lithium and iron were co-leached, iron was typically recovered as $FePO_4$ by adjusting the pH using ammonia (NH_4OH) or sodium hydroxide (NaOH).[42]

Building on this, studies have placed increasing emphasis on selective lithium leaching, aiming to leach lithium while

Table 1 Overview of hydrometallurgical approaches for recovery of metal compounds from EoL LFP batteries

Leaching step							Separation step		
Lixiviants		Additives	Temp. (°C)	Time (h)	S/L	LE_i (%)	Methods	Output	Ref.
Inorganic acid	0.3 M H_2SO_4	H_2O_2 (/Li molar ratio 2.07)	60	2	10.5 mL g^{-1}	96.85% Li 0.027% Fe 1.95% P	Li precipitation with Na_3PO_4 at 65 °C for 2 h	Li_3PO_4 (SE^b = 95.56%)	32
	2.5 M H_2SO_4	—	60	4	10 mL g^{-1}	97.2% Li 98.5% Fe	Li precipitation with Na_3PO_4 Fe precipitation with ammonia (pH ~2)	Li_2CO_3, $FePO_4·2H_2O$	33
	0.6 M H_2SO_4	1.3 MPa O_2	120	1.5	—	97.82% Li 0.022% Fe	Fe precipitation with 1 M NaOH (pH 5) Li precipitation with Na_3PO_4 at 95 °C for 0.5 h	Li_3PO_4 (SE = 97.97%) $FePO_4·2H_2O$ $Na_2SO_4·10H_2O$	34
	2 M H_2SO_4	Ascorbic acid	70	2	20 : 1	96.67% Li 93.25% Fe	—	—	35
	0.5 M H_3PO_4	—	RT	1	40 mL g		Refluxed at 85 °C for 9 h	LiH_2PO_4 (SE = ~75%) $FePO_4·2H_2O$	36
	2 M HCl	4 vol% H_2O_2	60	0.5	20 mL g^{-1}	>98% Li ~0.05% Fe	Fe precipitation with NaOH (pH 4) Li precipitation with Na_3PO_4 at 65 °C for 1 h	Li_3PO_4 (SE = >95%) $FePO_4$	37
	6.5 M HCl	15 vol% H_2O_2	60	2	1 : 5	92.15% Li	Fe^{3+} ions are selectively recovered as $FeCl_3$	Li_3PO_4 (SE = 93.68%)	38
						91.73% Fe	Li precipitation with Na_3PO_4 (pH 7) and 65 °C for 1 h	$FeCl_3$ (SE = 93.10%)	
Organic acid[a]	0.8 M AA	6 vol% H_2O_2	50	0.5	120 g L^{-1}	95.05% Li >1% Fe, Al	Li precipitation with Na_2CO_3 at 95 °C for 1 h	Li_2CO_3 (purity = 99.95%) $FePO_4$	39
	0.8 M FA	8 vol% H_2O_2	60	1	50 g L^{-1}	99.96% Li <0.1% Fe	Li precipitation with Na_2CO_3 at 95 °C	Li_2CO_3 (SE = 85.05%) $FePO_4$	40
	2.5 M FA	0.12 L min^{-1} O_2	70	3	25 mL g^{-1}	99.9% Li 1.7% Fe	—	$FePO_4$	41
	4 M MSA	18 vol% H_2O_2	RT	1.5	80 g L^{-1}	94% Li 95% Fe	Li precipitation with Na_2CO_3 at 96 °C Fe precipitation with H_2O_2 (pH 4)	Li_2CO_3 (SE = 99%) $FePO_4$	42
Oxidant	pH 3.5 H_2O	600 mL min^{-1} air	RT	5	10 mL g^{-1}	99.3% Li 0.02% Fe, P	Li precipitation with Na_2CO_3 at 95 °C	Li_2CO_3 $FePO_4$	43
	pH 7 H_2O	2.7 M H_2O_2	RT	4	5 : 1	95.4% Li 0.05% Fe	—	$FePO_4$	44

[a] AA, FA, and MSA stand for acetic acid, formic acid, and methanesulfonic acid, respectively. [b] SE stands for separation efficiency of recovered compounds, *e.g.*, Li_3PO_4, Li_2CO_3, $FePO_4$, *etc.*

minimizing the co-dissolution of iron and phosphorus. In the presence of an oxidizing agent such as H_2O_2 or O_2, the E_h increases, promoting the oxidation of Fe^{2+} to Fe^{3+}, which subsequently precipitates as solid $FePO_4$ (Fig. S1). This shifts the chemical equilibrium toward the Li^+ and $FePO_4$ domain, thereby enabling selective leaching of lithium while retaining iron in the solid phase, as represented by the following reaction:[13]

$$\begin{gathered}2LiFePO_4(s) + H_2SO_4(aq) + H_2O_2(aq) \rightarrow \\ Li_2SO_4(aq) + 2FePO_4(s) + 2H_2O(l).\end{gathered} \quad (2)$$

The above redox-governed leaching mechanisms highlight the importance of precisely controlling E_h and pH through appropriate combinations of lixiviants and oxidants to achieve high lithium extraction efficiency while minimizing the co-dissolution of iron.[28] This selectivity is particularly critical not only for reducing reagent consumption during downstream separation, but also for preserving $FePO_4$ as a solid-phase precursor suitable for cathode material regeneration. For example, Li *et al.*[32] used H_2SO_4 with H_2O_2 as an oxidant to leach over 96% lithium, while Fe and P were leached only 0.027% and 1.95%, respectively, under the optimal conditions (0.3 M H_2SO_4, 2.07 molar ratio of H_2O_2/Li, 60 °C, 2 h, and S/L = 10.5 mL g^{-1}). Similarly, Li *et al.*[40] employed 0.8 M formic acid (HCOOH) and 8 vol% H_2O_2 at 60 °C to achieve 99.9% LE_{Li} and only 0.05% LE_{Fe}. Zhao *et al.*[41] reported one of the highest Li/Fe separation coefficients (>21 000) using a HCOOH and oxygen (O_2) system, demonstrating exceptional control over redox and pH conditions in selective leaching. Additionally, mechanochemical activation, as reported in citric acid ($C_6H_8O_7$)[46] and oxalic acid ($C_2H_2O_4$)[47] systems, further enhances leaching kinetics under mild or even room-temperature conditions.

Following the leaching step, lithium is typically recovered *via* precipitation as Li_2CO_3 and Li_3PO_4 depending on the process design, according to the following reactions:[48,49]

$$2Li^+(aq) + Na_2CO_3(aq) \rightarrow Li_2CO_3(s) + 2Na^+(aq) \quad (3)$$

$$3Li^+(aq) + Na_3PO_4(aq) \rightarrow Li_3PO_4(s) + 3Na^+(aq). \quad (4)$$

The precipitation of lithium from the leachate is governed by solubility equilibria, where lithium reacts with appropriate anions to form sparingly soluble compounds. For example, Li_2CO_3 and Li_3PO_4 have solubilities of about 1.3 g per 100 mL and 0.04 g per 100 mL in water at 20 °C, respectively. These low solubilities enable selective recovery of lithium as solid products from aqueous leachates. The effectiveness of this step depends on several parameters, including pH, temperature, and concentration. These principles form the basis of many reported processes and have been successfully applied in the practical lithium recovery step. For instance, precipitation of Li_2CO_3 by adding Na_2CO_3 is widely used,[50–52] and Na_3PO_4 has been used to precipitate Li_3PO_4 at temperatures ranging from 65 to 95 °C within 2 h.[49,53,54] Iron is often recovered as $FePO_4$ generated after selective lithium leaching.[34,39,40,47] To reduce sodium contamination, CO_2 gas purging is also widely employed to recover lithium as Li_2CO_3 instead of using Na_2CO_3.[55,56] The purity of these recovered compounds typically exceeds 99% separation efficiency (SE_i; i = metal compound), enabling their direct reuse in cathode material synthesis.[57–60]

As a next step, a number of hydrometallurgical studies have extended beyond the recovery of metal compounds by demonstrating the synthesis of S-LFP using the as-recovered metal compounds.[27,35,59] This integration step is critical for the realization of a closed-loop LiB system (Fig. 1a), wherein EoL cathode materials are converted back into functional precursors and subsequently reused in new battery manufacturing.[59] The as-recovered $FePO_4$ has been reacted with recovered Li_2CO_3 in solid-state or hydrothermal processes, sometimes aided by reducing agents such as glucose ($C_6H_{12}O_6$) or carbon black as a conductive agent.[61–63] The optimized synthesis temperatures typically range from 600 °C to 750 °C, depending on precursor crystallinity and synthesis routes.[13,64] In most cases, the electrochemical performance of S-LFP is comparable to that of commercial-grade materials. For example, Zheng *et al.*[33] reported an initial discharge capacity of 155.4 mAh g^{-1} at 0.1C for S-LFP synthesized from recovered $FePO_4 \cdot 2H_2O$ and Li_2CO_3, which is close to the theoretical capacity (170 mAh g^{-1}) of the LFP battery. Similarly, Song *et al.*[35] demonstrated that S-LFP microsheets prepared hydrothermally using leachate-derived Li^+ and Fe^{3+} achieved >98% capacity retention over 100 cycles at 1C, indicating excellent structural stability. In some cases, the use of hierarchical or nanostructured $FePO_4$ precursors even improved rate capability compared to pristine LFP, due to enhanced lithium-ion diffusion.[65,66] These findings collectively validate the feasibility of full-loop LFP regeneration using recovered materials from spent cathodes. Moreover, the comparable electrochemical properties of S-LFP confirm that hydrometallurgical recycling can not only close the material loop but also meet the performance standards required for modern LFP applications.

While the preparation of S-LFP cathodes using as-recovered metal compounds represents a meaningful step toward material circularity,[67–69] it does not fully address the core challenges associated with conventional hydrometallurgical recycling. Chief among these is the generation of substantial volumes of wastewater containing residual acids, salts, and metal ions, which require further treatment or disposal, thereby increasing both environmental impact and operational cost.[24,26,70,71] To achieve truly sustainable and scalable hydrometallurgical recycling of LFP batteries, future research must focus on minimizing chemical agent consumption through closed-loop reuse of wastewater to enhance economic feasibility. Ultimately, the integration of metal recovery with environmentally benign and cost-effective strategies remains a critical challenge and opportunity for next-generation battery recycling systems.

3. Closed-loop hydrometallurgical approaches by wastewater reuse

This review provides an in-depth analysis of studies that have reused aqueous effluents generated after the hydrometallurgical recovery of metals from EoL LFP batteries in subsequent leaching, separation, or even pre-treatment steps. The focus on aqueous waste streams stems from the fact that the reuse of organic solvents or hydrophobic ionic liquids following solvent extraction has already been extensively reported in the literature,[72–76] and is therefore beyond the scope of this work. In addition, although numerous studies have demonstrated the preparation of S-LFP materials using as-recovered metals, the electrochemical performance of these regenerated materials is not addressed here. Instead, this review highlights various strategies and outcomes aimed at minimizing environmental impact and improving the economic feasibility of hydrometallurgical processes through the effective reuse of wastewater. Since the chemical composition of wastewater varies significantly depending on the reagents used, relevant studies are categorized and analyzed based on the type of lixiviant used during the initial leaching stage.

3.1 Inorganic acids

3.1.1 Phosphoric acid. H_3PO_4, a relatively weak inorganic acid compared to strong acids such as H_2SO_4, is often used as a lixiviant in LFP hydrometallurgical recycling because it partially matches the chemical composition of LFP.[77,78] Similar to other inorganic acids, H_3PO_4 can also selectively leach only Li in the presence of oxidants, such as H_2O_2. After the leaching step, Li can be simply recovered as Li_3PO_4 from the PO_4^{3-}-containing leachate *via* heating or evaporation without adding other agents, reducing chemical consumption. The chemical reaction between LFP and H_3PO_4, with and without H_2O_2, can be represented by the following reactions:

$$\begin{aligned}\mathrm{LiFePO_4(s) + H_3PO_4(aq)} &\rightarrow \mathrm{LiH_2PO_4(aq) + Fe^{3+}(aq)} \\ &\quad + \mathrm{PO_4{}^{3-}(aq)(pH < 2)}\end{aligned} \tag{5}$$

$$\begin{aligned}&\mathrm{2LiFePO_4(s) + 2H_3PO_4(aq) + H_2O_2(aq)} \rightarrow \\ &\mathrm{2LiH_2PO_4(aq) + 2FePO_4(s) + 2H_2O(l).}\end{aligned} \tag{6}$$

Fe^{3+} precipitates almost entirely as $FePO_4$ at pH 2 and above.[79] Given that the pH value of 0.1 M H_3PO_4 is about 1.56, using H_3PO_4 alone as a lixiviant will primarily leach lithium and a small amount of iron.

Chen *et al.*[80] proposed a scalable and simplified closed-loop hydrometallurgical process for recovering valuable metals from a mixed stream of EoL LiBs, including LFP, NCM, and LMO cathodes (Fig. 3). In this study, H_3PO_4 served as a lixiviant, while H_2O_2 acted both as a reductant (for Co^{3+} and Mn^{4+}) and an oxidant (for Fe^{2+}). Optimal leaching conditions (2 M H_3PO_4, 4 vol% H_2O_2, 60 °C, 1 h, S/L = 20 mL g^{-1}) enabled near-complete leaching of lithium, nickel, cobalt and manganese (≥95.5% LE), with iron remaining as insoluble $FePO_4$.[80] The subsequent addition of oxalic acid led to the co-precipitation of nickel, manganese and cobalt as their oxalates, while lithium remained in the solution and was later recovered as Li_2CO_3 *via* precipitation with NaOH and Na_2CO_3. Importantly, during co-precipitation, H_3PO_4 was regenerated *in situ* due to the stronger acidity of oxalic acid, and this regenerated acid was reused for up to five leaching cycles without significant loss in LE_i (>90% for lithium and >70% for nickel, manganese, and cobalt) recovery per cycle, demonstrating strong reagent stability and process reusability. From an economic standpoint, recycling 1 kg of mixed EoL LiB cathodes generated an overall profit of $26.79, thanks to the low consumption of reagents, energy-efficient operation, and elimination of waste-

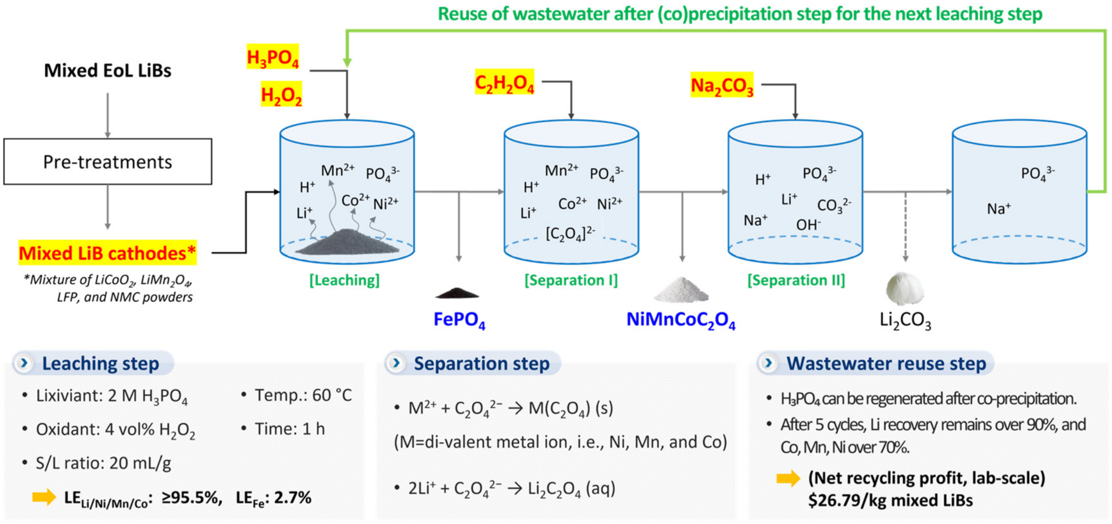


Fig. 3 Conceptual closed-loop hydrometallurgical process for recovery of $FePO_4$, $NiMnCoC_2O_4$, and Li_2CO_3 from mixed LiB cathode materials using H_3PO_4 and H_2O_2 as leaching agents. Drawn by the authors based on data reported in ref. 80 (hereinafter, the dashed line in each figure indicates that the information is not given in detail or was omitted in the referred study).

water treatment.[78] However, some practical limitations were noted. Specifically, although 1.14 L of H_3PO_4 was regenerated, the actual volume required for leaching 420 g of cathode material at the optimal S/L ratio (20 mL g^{-1}) would be 8.4 L, suggesting that regenerated H_3PO_4 constitutes only ~14 vol% of the total lixiviant demand. The study does not explicitly address the concentration and purity of the regenerated acid, nor quantify the amount of fresh H_3PO_4 needed per cycle, which are critical for scaling up the process. Furthermore, the potential accumulation of Na^+ or oxalate-derived impurities over multiple cycles was not investigated, leaving uncertainties regarding the long-term stability of the leaching process. In particular, the influence of Na^+ buildup in the recycled solution on subsequent LE_{Li} warrants further examination. Additionally, a quantitative assessment of the concentration of regenerated H_3PO_4 and the amount of fresh acid required in each cycle would provide valuable insights for optimizing future closed-loop hydrometallurgical designs. Finally, although the study demonstrates the *in situ* regeneration and reuse of H_3PO_4 over five leaching cycles, the reported economic analysis does not account for the potential cost savings associated with this reuse. In particular, the calculation excludes the benefit of eliminating wastewater treatment for spent acid. Despite these limitations, the process demonstrates strong potential for sustainable metal recovery and cathode regeneration, in line with green chemistry and circular economy objectives.

Guo *et al.*[69] also developed an eco-friendly, closed-loop hydrometallurgical process for recovering metals from EoL LFP cathodes using a low-solvent solid-state reaction (LSR), as shown in Fig. S2. After discharging the batteries in 1 M NaCl, LFP powders were leached with H_3PO_4 and 12.5 vol% H_2O_2 at 30 °C for 3 h (S/L = 1 : 3 g mL^{-1}, $H_3PO_4 : Li$ (M) = 1 : 1), resulting in complete decomposition into $FePO_4$ and LiH_2PO_4. Excess PO_4^{3-} was removed by adding $CaCl_2$, producing $Ca(H_2PO_4)_2$ as a recoverable byproduct, and lithium was precipitated as high-purity Li_2CO_3 using Na_2CO_3 at 90 °C for 2 h.[69] A novel aspect of this work is the reuse of the final NaCl-based wastewater for battery discharging, thereby minimizing fresh reagent use. However, the exact volume and ionic composition of the recycled NaCl solution were not provided. Without information on the accumulation of ions, *e.g.*, Na^+, Ca^{2+}, Cl^-, PO_4^{3-}, or $H_2PO_4^-$ over multiple cycles, it is difficult to evaluate the long-term chemical stability of the discharge solution or its impact on battery safety and discharging efficiency. Furthermore, the effects of impurities other than NaCl on battery discharging and reuse performance were not addressed and warrant further investigation. Economically, the LSR-based process yielded a net profit of \$2.06 per kg of EoL LFP powder, primarily due to elimination of wastewater treatment. However, the study lacks a sensitivity analysis for multi-cycle reuse and does not quantify the long-term economic benefits associated with NaCl recycling. Nonetheless, this study demonstrates a promising low-temperature and closed-loop strategy for LFP recycling, with integrated wastewater reuse and product recovery.

Yang *et al.*[67] developed a closed-loop hydrometallurgical process to recover battery-grade $FePO_4 \cdot 2H_2O$ from delithiated LFP cathodes by reusing evaporated wastewater in successive leaching cycles (Fig. S3). Leaching was performed with 6 M H_3PO_4 at 75 °C for 3 h (S/L = 133.3 g L^{-1}), achieving 99.2% LE_{Fe} and 98.0% LE_P. Trace Cu was removed as CuS using $Na_2S \cdot 9H_2O$, and $FePO_4 \cdot 2H_2O$ was recovered by crystallization at 90 °C for 4 h with a dilution ratio of 3.5, yielding a 93.5% $SE_{FePO_4 \cdot 2H_2O}$. Importantly, the study implemented a closed-loop approach by reusing the mother liquor from the crystallization step as a lixiviant after simple evaporation and concentration. This reuse strategy was applied over three leaching cycles, during which LE_{Fe} remained above 99%, and the $SE_{FePO_4 \cdot 2H_2O}$ exceeded 93%, indicating sufficient operational stability and reusability of the wastewater.[67] This process effectively minimized wastewater discharge, as the recycled solution was used to dilute the fresh H_3PO_4 input. However, the available data are insufficient to fully validate a truly closed-loop process. For instance, approximately 7% of Fe remained in the crystallization liquor, suggesting non-negligible iron losses. In addition, Na^+ ions introduced during copper removal may gradually accumulate with each cycle. To establish a precise material and mass balance, it is essential to quantify impurity concentrations, particularly residual metal ions and Na^+, in each leaching cycle relative to the original solution. It would also be meaningful to investigate how many cycles are required before these accumulated impurities begin to adversely affect metal recovery efficiency. Furthermore, the study does not report key variables such as the final H_3PO_4 concentration or the total volume of the concentrated wastewater, making it difficult to assess how much the recycled solution contributes to the following leaching steps. Despite these limitations, the reported methodology offers meaningful insight into the development of closed-loop hydrometallurgical systems. Finally, a techno-economic analysis was conducted. Although the study did not quantify cost savings from wastewater reuse as a function of cycle number, it estimated that recycling 1 ton of delithiated LFP cathode could yield 990 kg of $FePO_4 \cdot 2H_2O$, generating \$1534.5 in revenue. With total operational costs (including energy, labor, and equipment) of \$643.0, the resulting net profit was \$891.5 per ton, equivalent to \$0.89 per kg, highlighting the commercial potential of this process for sustainable LFP battery recycling.

3.1.2 Sulfuric acid.

In LiB recycling, H_2SO_4 is commonly used as a lixiviant due to its strong acidity, low cost, and ability to effectively dissolve a wide range of metal compounds, particularly lithium and transition metals.[81] It facilitates high metal recovery rates under relatively mild conditions and is compatible with various oxidants, making it suitable for scalable and efficient hydrometallurgical processes. For LFP batteries, sulfuric acid is often combined with H_2O_2 to enable the selective leaching of lithium as represented by reaction (2).

The study by Tao *et al.*[82] presents a practical and environmentally sustainable hydrometallurgical method for recycling EoL LFP batteries, utilizing cathode scrap obtained through disassembly as the primary feedstock (Fig. 4). Unlike conven-

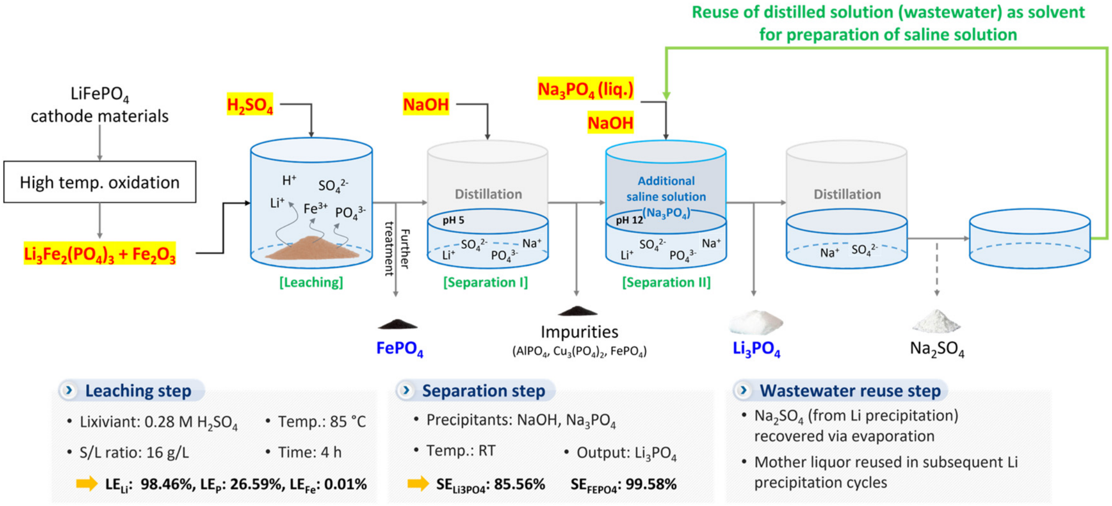


Fig. 4 Conceptual closed-loop hydrometallurgical process for recovery of $FePO_4$ and Li_3PO_4 from oxidized LFP cathode materials using H_2SO_4 as a lixiviant. Drawn by the authors based on data reported in ref. 82.

tional oxidative leaching systems, this method involves high-temperature pre-treatment of LFP powder in air at 600 °C to convert it into $Li_3Fe_2(PO_4)_3$ and Fe_2O_3, which are more readily leached under mild acidic conditions. Under optimized leaching conditions (0.28 M H_2SO_4, 85 °C, 4 h, $Li:H_2SO_4$ molar ratio = 2.63), the process achieved 98.46% LE_{Li}, with 0.01% LE_{Fe} and 26.59% LE_P. Lithium was subsequently recovered as Li_3PO_4 with 85.56% SE, while the leaching residue was thermally treated to produce $FePO_4$ with a 99.58% SE. A key feature of this process is the reuse of the Na_2SO_4-rich wastewater generated after Li_3PO_4 precipitation. Instead of discarding it, the Na_2SO_4 solution was directly reused as a preparation solution for the precipitant in subsequent cycles. The salt remained effective over multiple reuse cycles without significant performance loss, thereby minimizing the need for fresh reagent input and reducing waste generation.[82] This strategy effectively realized a partially closed-loop system and markedly improved both the sustainability and cost-efficiency of the process. Preliminary economic analysis revealed favorable profit margins, primarily attributed to the reuse of Na_2SO_4, elimination of costly oxidants, and the high yield of recoverable products. Moreover, the process demonstrated potential for industrial scale-up with reduced chemical consumption and minimal environmental burden. Nevertheless, several challenges remain in achieving a fully closed-loop operation. The long-term chemical stability of recycled Na_2SO_4 and the impact of impurity accumulation on Li_3PO_4 crystallization were not evaluated. In addition, the fate of partially dissolved phosphorus (LE_P = 26.59%) remains unclear, raising the potential for phosphorus accumulation over multiple cycles. The mass balance of sulfuric acid usage was also not addressed, leaving uncertainties about its regeneration or net consumption. Furthermore, process efficiency may be limited by design parameters such as pulp density. Notably, the leaching was conducted at a very low pulp density of 16 g L^{-1}, which resulted in a large volume of leachate per unit mass of solid. As a consequence, several distillation steps were required to concentrate lithium to a level sufficient for downstream recovery.

Beyond the reuse of Na_2SO_4 as a preparation solution for the precipitant, recent studies have explored bipolar membrane electrodialysis (BMED) as a promising approach to convert Na_2SO_4-containing wastewater into two valuable reagents, *i.e.*, H_2SO_4 and NaOH.[25,83–88] Under an applied electric field, Na^+ and SO_4^{2-} ions are selectively transported across the bipolar membrane stack, generating H_2SO_4 in the acid compartment and NaOH in the base compartment. These regenerated reagents can be directly reused in the same hydrometallurgical process, *i.e.*, H_2SO_4 as a lixiviant and NaOH as a pH modifier, thereby reducing chemical consumption and secondary waste production. This approach further supports the development of a fully closed-loop, resource-efficient recycling system for LFP batteries. However, its practical implementation requires careful evaluation of BMED conversion efficiency and associated energy consumption,[84,89,90] as these factors directly impact the overall sustainability and economic feasibility of the closed-loop system.

3.2 Inorganic acidic salt-based lixiviants

3.2.1 Sodium phosphate monobasic. NaH_2PO_4 serves as an effective lixiviant for LFP due to its ability to provide protons (H^+) under mildly acidic conditions.[22,30] When combined with an oxidant, it enables the selective leaching of lithium ions while stabilizing iron as solid $FePO_4$ as represented by the following reaction:

$$\begin{aligned}&2LiFePO_4(s) + NaH_2PO_4(aq) + H_2O_2(aq)\\&\rightarrow 2FePO_4(s) + Li_2NaPO_4(aq) + 2H_2O(l).\end{aligned} \quad (7)$$

Zhou *et al.*[91] developed a mild and closed-loop hydrometallurgical process for selectively recovering metals from EoL LFP cathodes using NaH_2PO_4 and H_2O_2 as leaching agents (Fig. 5).

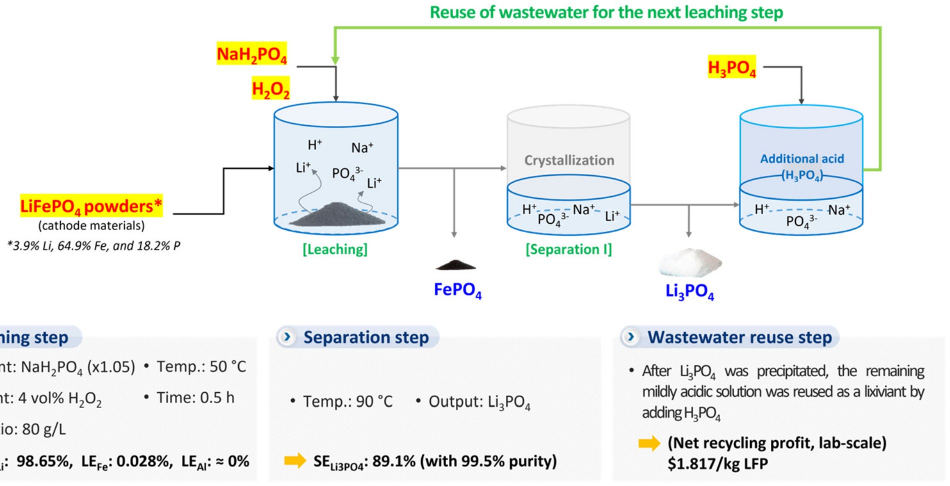


Fig. 5 Conceptual closed-loop hydrometallurgical process for recovery of $FePO_4$ and Li_3PO_4 from LFP cathode materials using NaH_2PO_4 and H_2O_2 as leaching agents. Drawn by the authors based on data reported in ref. 91.

Under optimized conditions (1.05 times theoretical molar amount of NaH_2PO_4, 4 vol% H_2O_2, 50 °C, 30 min, S/L = 80 g L^{-1}), lithium was leached with 98.65% efficiency, while iron dissolution remained as low as 0.028%. Lithium was recovered as Li_3PO_4 *via* evaporative crystallization with 99.5% purity and 89.1% SE.[91] According to the authors, approximately 92% of water was evaporated during this process, concentrating the remaining 8% into a Li_3PO_4-saturated solution suitable for crystallization. Importantly, the process integrates a closed-loop design by reusing the post-crystallization brine. By simply adding H_3PO_4, the brine was re-acidified and directly reused as a leaching agent for the next cycle, avoiding the need for neutralization or effluent treatment. This not only reduces initial lixiviant consumption but also minimizes wastewater treatment costs. Economically, the process yields $1.817 profit per kg of EoL LFP cathode powder, based on the recovery of 0.75 kg $FePO_4$ and 0.19 kg Li_3PO_4 per kilogram of feed. The cost-effectiveness arises from simple brine recycling and the elimination of costly wastewater treatment steps. Although the study presents a promising closed-loop hydrometallurgical process for LFP recycling, several aspects remain unaddressed for it to be considered a fully closed-loop process. First, the composition and chemical characteristics of the reused leachate, such as pH and impurity content, were not thoroughly analyzed, making it unclear how the regenerated solution performs over multiple cycles. Additionally, the potential accumulation of Na^+ and other unreacted metal impurities in the recycled brine was not evaluated, which could negatively affect lithium recovery efficiency or product purity in long-term operation. The study also does not specify the proportion of fresh reagents, such as NaH_2PO_4 or H_3PO_4, required in each reuse cycle, making it difficult to assess the actual reagent savings and sustainability of the system. Moreover, no long-term cycling tests were performed, and critical metrics such as water and energy consumption or secondary waste generation were omitted.

3.2.2 Sodium hydrogen sulfate.

$NaHSO_4$ serves as a mild leaching agent that enables selective lithium extraction from LFP when combined with H_2O_2, while minimizing iron dissolution.[27] It also supports a closed-loop process, as the resulting Na_2SO_4 can be regenerated into $NaHSO_4$ using H_2SO_4, reducing reagent consumption and wastewater generation. The selective leaching reaction between LFP and $NaHSO_4$ in the presence of H_2O_2 can be represented by the following chemical reaction:

$$\begin{aligned}&2\mathrm{LiFePO_4(s)} + 2\mathrm{NaHSO_4(aq)} + \mathrm{H_2O_2(aq)} \rightarrow \\ &\quad 2\mathrm{LiNaSO_4(aq)} + 2\mathrm{FePO_4(s)} + 2\mathrm{H_2O(l)}.\end{aligned} \tag{8}$$

The study conducted by Gong *et al.*[27] presents a sustainable and highly efficient closed-loop hydrometallurgical process for recovering lithium from EoL LFP batteries (Fig. 6). After pretreatment, including Na_2SO_4-based discharge, manual disassembly, and ultrasonic stripping, the purified LFP powders were leached using $NaHSO_4$ with H_2O_2. Under optimized conditions (1.1 times theoretical amount of $NaHSO_4$, 2 vol% H_2O_2, 65 °C, 15 min, S/L = 100 g L^{-1}), the process achieved 99.84% LE_{Li}, with only 0.048% LE_{Fe}. Lithium was then recovered as high-purity Li_2CO_3 with 99.5% purity *via* supersaturated Na_2CO_3 precipitation at 95 °C.[27] A key innovation of this work is the regeneration of $NaHSO_4$ from the Na_2SO_4-rich wastewater produced after lithium precipitation. By adding H_2SO_4, Na_2SO_4 was effectively converted back into $NaHSO_4$, which was reused in subsequent leaching cycles. This reagent regeneration step eliminates wastewater treatment and significantly reduces chemical consumption, enhancing both environmental and economic viability. However, despite the strong performance and closed-loop design, several aspects must be further addressed to realize a truly complete closed-loop system. First, the study does not report the mass balance and regeneration efficiency of $NaHSO_4$ over multiple cycles, leaving uncertainty about the self-sufficiency of the lixiviant.

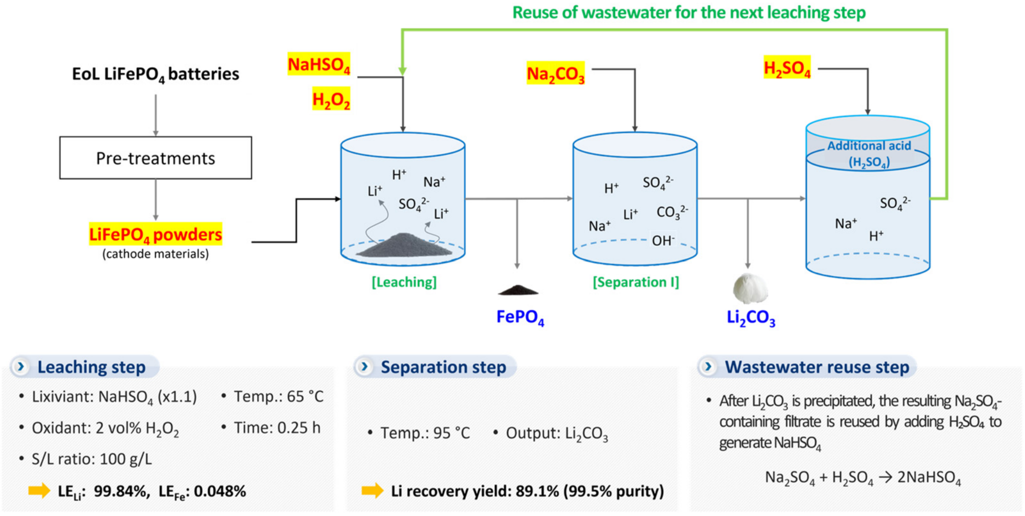


Fig. 6 Conceptual closed-loop hydrometallurgical process for recovery of FePO$_4$ and Li$_2$CO$_3$ from LFP cathode materials using NaHSO$_4$ and H$_2$O$_2$ as leaching agents. Drawn by the authors based on data reported in ref. 27.

Second, the long-term reuse of regenerated NaHSO$_4$ and its effect on LE and product purity has not been tested beyond a single cycle. Third, despite the possibility that accumulation of impurities such as Na$^+$, SO$_4^{2-}$, or residual carbonates from repeated use of reagents may interfere with lithium crystallization, these effects have not been systematically evaluated. Additionally, no quantitative data are provided on how much fresh H$_2$SO$_4$ input is still required for the next leaching cycle, making it difficult to assess the degree to which external chemical inputs can be minimized.

3.3 Inorganic oxidizing salt-based lixiviants

3.3.1 Ferric sulfate (Fe$_2$(SO$_4$)$_3$). Fe$_2$(SO$_4$)$_3$ is used as a lixiviant due to its dual role as a mild acid and an effective oxidant.[92] In aqueous solution, Fe^{3+} ions facilitate the oxidative dissolution of target metals by converting them into soluble ionic forms, while simultaneously maintaining a mildly acidic environment (about pH 2–3) without the need for strong acids. This enables efficient leaching of lithium from LFP under mild conditions. Compared to conventional strong acids, Fe$_2$(SO$_4$)$_3$ offers lower corrosivity and environmental impact, making it a more sustainable option for hydrometallurgical processes. In LFP recycling, the chemical reaction can be described as follows:

$$6\mathrm{LiFePO_4(s)} + \mathrm{Fe_2(SO_4)_3(aq)} + 3\mathrm{H_2O_2(aq)} \rightarrow 3\mathrm{Li_2SO_4(aq)} + 6\mathrm{FePO_4(s)} + 2\mathrm{Fe(OH)_3(s)}. \quad (9)$$

The study by Niu *et al.*[93] presents a highly integrated and sustainable closed-loop hydrometallurgical process, employing a coupling strategy of leaching and solvent extraction (Fig. 7). In this system, Fe$_2$(SO$_4$)$_3$ acted as both the lixiviant and oxidant, with H$_2$O$_2$ promoting Fe^{2+} oxidation. Under optimized conditions (Fe$_2$(SO$_4$)$_3$: LFP (M) = 1 : 6, H$_2$O$_2$ = 0.4 mL g^{-1} LFP, 40 °C, 30 min, S/L = 300 g L^{-1}), LE$_{Li}$ reached 99.41%, and 96% of FePO$_4$ was retained in the solid phase. It is worth noting that, since H$_2$SO$_4$ and H$_2$O$_2$ were co-introduced dropwise during leaching, the resulting acidic environment likely facilitated the partial dissolution of Fe(OH)$_3$ into the leachate, rather than allowing complete precipitation as a solid. Then, Fe^{3+} was selectively removed from the leachate using a solvent system of 40 vol% N235 (tri(octyl-decyl)amine) and 15 vol% TBP (tributyl phosphate) in sulfonated kerosene, achieving 97.22% extraction in a four-stage setup. Stripping with 1.4 M H$_2$SO$_4$ recovered Fe^{3+}, and Li$_2$CO$_3$ was precipitated from the purified raffinate at 80 °C with over 99% purity.[93] A major innovation of this work is not only the regeneration of the organic phase, but also the possibility of the regeneration of the stripped aqueous phase for the next leaching cycle. The Fe-loaded extractant was restored in two steps: water scrubbing to remove acidity and NaOH saponification to regenerate extractant activity. The regenerated solvent maintained Fe^{3+} extraction performance over 13 cycles without loss in phase separation or efficiency. Additionally, the stripped liquor was proposed for reuse in the reformation of Fe$_2$(SO$_4$)$_3$ as a lixiviant, and this strategy was further validated through experiments, achieving over 99% lithium recovery (both leaching and separation) in the coupled leaching system. An economic analysis estimated a net cost of \$3.49 per kg of LFP, demonstrating commercial viability. Despite these promising results, several limitations remain. Minor Fe^{3+} losses per cycle suggest the need for periodic replenishment, and the effects of impurity accumulation on Li$_2$CO$_3$ purity and long-term process stability were not assessed.

3.3.2 Sodium persulfate. Na$_2$S$_2$O$_8$ serves as a powerful oxidant for the selective leaching of lithium from EoL LFP batteries.[28,29] It enables the oxidation of Fe^{2+} in the LFP structure to Fe^{3+}, forming insoluble FePO$_4$ while releasing lithium ions into solution. This reaction mimics the electrochemical charging process and preserves the olivine structure of FePO$_4$,

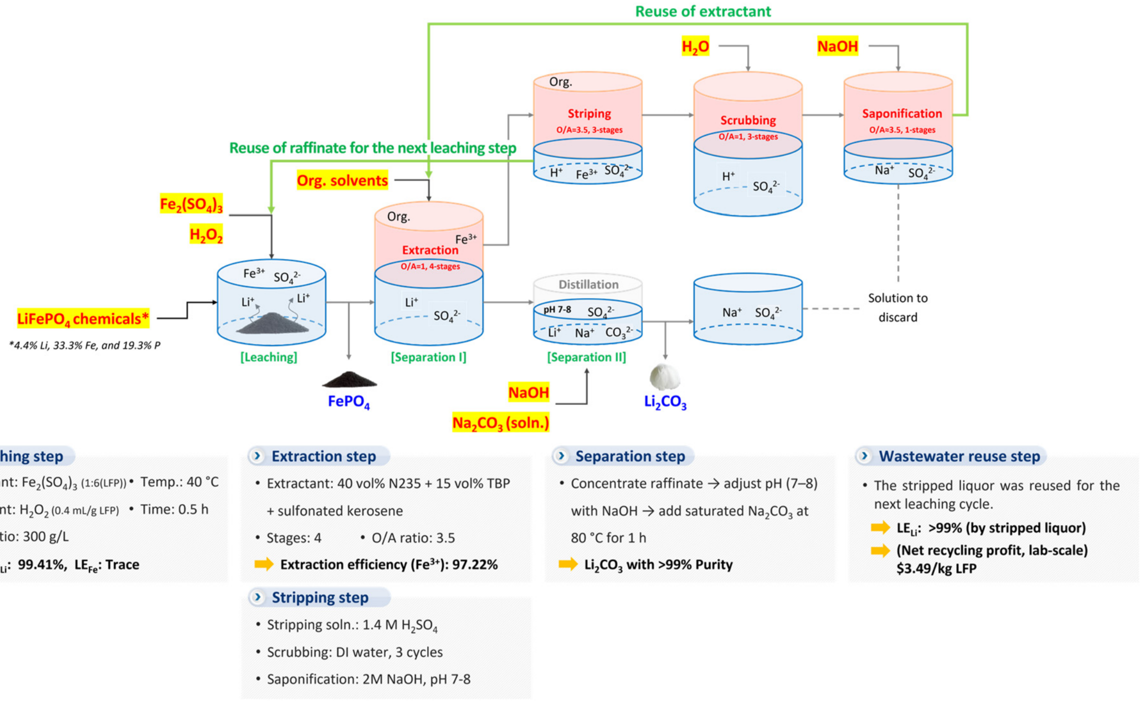


Fig. 7 Conceptual closed-loop hydrometallurgical process for recovery of $FePO_4$ and Li_2CO_3 from pure LFP chemicals using $Fe_2(SO_4)_3$ and H_2O_2 as leaching agents; O/A stands for the ratio of the organic phase to the aqueous phase. Drawn by the authors based on data reported in ref. 93.

thereby facilitating solid–liquid separation. The overall reaction can be represented as follows:

$$2LiFePO_4(s) + Na_2S_2O_8(aq) \rightarrow 2FePO_4(s) + Li_2SO_4(aq) + Na_2SO_4(aq). \tag{10}$$

Zhang *et al.*[28] developed a sustainable closed-loop hydrometallurgical process for LFP recycling with high lithium selectivity and minimal waste generation by using $Na_2S_2O_8$ (Fig. 8). Under optimized conditions (1.05 times theoretical amount of $Na_2S_2O_8$, 25 °C, 20 min, S/L = 300 g L^{-1}), LE_{Li} reached 99.9% with 0.048% LE_{Fe}. Then, lithium was subsequently recovered as high-purity Li_2CO_3 (over 99% purity) by adjusting the leachate pH to 7.5–8.5 using NaOH, followed by the addition of saturated Na_2CO_3 at 95 °C for 2 h. A key feature of this study is the closed-loop recycling system, wherein the mother liquor after lithium precipitation is recycled back into the leaching process, reducing the need for fresh reagents and minimizing waste production. This efficient reuse of materials significantly enhances the economic viability of the process, with an estimated net profit of $510.8 per ton of EoL LFP cathode material.[28] These results highlight the strong potential for industrial-scale application of this method in sustainable LFP battery recycling.

To further support reagent circularity, $Na_2S_2O_8$ can be directly regenerated from Na_2SO_4-rich solutions *via* anodic electrolysis, offering a closed-loop route for oxidant recycling. As demonstrated in a patent reported by Kimizuka *et al.*,[94] $Na_2S_2O_8$ could be regenerated with 99.8% purity and 82% current efficiency over 10 h of electrolysis at 6.6 V. This approach enables complete reuse of the oxidant without requiring fresh $Na_2S_2O_8$ in subsequent leaching steps, thereby further enhancing both the sustainability and economic feasibility of the process. Using the same method, it has been reported that ammonium persulfate ($(NH_4)_2S_2O_8$) could be regenerated *via* anodic electrolysis from ammonium sulfate-based wastewater obtained after lithium recovery, forming a closed-loop oxidant recycling pathway.[94,95] As reported by Shentu *et al.*,[95] $(NH_4)_2S_2O_8$ can be produced through electrolysis, enabling repeated use of the oxidant without requiring fresh addition in each cycle. Although the reuse of $(NH_4)_2S_2O_8$ as a lixiviant for LFP recycling has been conceptually proposed, it was only introduced theoretically in some studies and not experimentally validated for reuse in actual closed-loop hydrometallurgical processes.

3.4 Inorganic alkaline lixiviant

3.4.1 Sodium hydroxide. NaOH functions as both a lixiviant and an *in situ* oxidant in the selective recovery of lithium from thermally treated LFP cathodes.[96] In this oxidizing alkaline system, NaOH facilitates the dissolution of lithium and phosphate into solution, while iron is oxidized from Fe^{2+} to Fe_3O_4. Concurrently, OH^- is reduced, generating H_2 gas, thereby eliminating the need for external oxidants. The chemical reaction can be represented as follows:

$$3LiFePO_4(s) + 6NaOH(aq) \rightarrow 3Li^+(aq) + 6Na^+(aq) + 3PO_4{}^{3-}(aq) + Fe_3O_4(s) + 2H_2O(l) + H_2(g). \tag{11}$$

Yang *et al.*[96] developed a novel and efficient closed-loop hydrometallurgical approach to recover lithium from EoL LFP

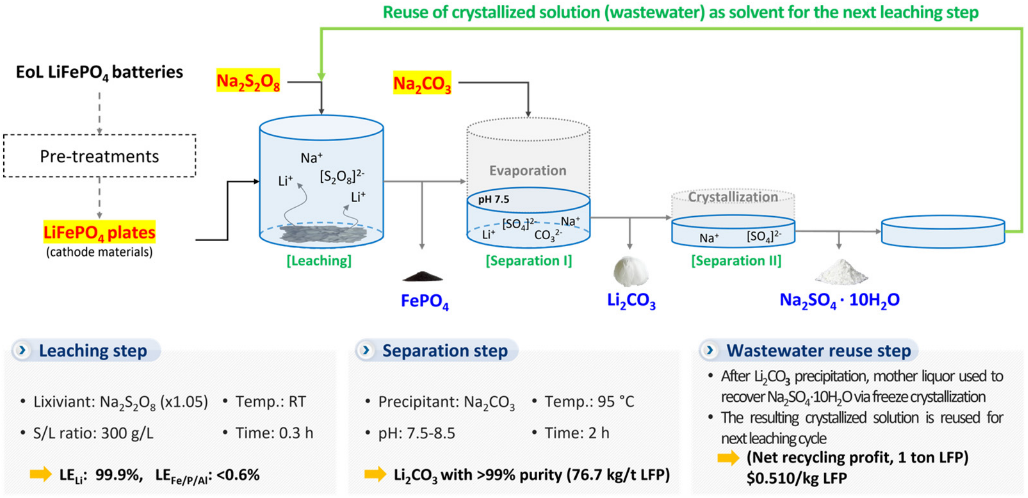


Fig. 8 Conceptual closed-loop hydrometallurgical process for recovery of $FePO_4$ and Li_2CO_3 from LFP cathodes using $Na_2S_2O_8$ as a lixiviant. Drawn by the authors based on data reported in ref. 28.

batteries, using cathode scrap as the main input material. In contrast to conventional acidic or oxidative leaching systems, this process uniquely employs NaOH as both the leaching agent and the oxidant, thereby eliminating the need for external oxidants (Fig. 9). Under optimal conditions (2 M NaOH, S/L = 1 : 50 g mL^{-1}, 50 °C, 2 h), lithium and phosphorus were leached at 98.2% and 99.9%, respectively, while iron remained as solid Fe_3O_4. Then, lithium was recovered as high-purity Li_3PO_4 *via in situ* precipitation at 90 °C.[96] A key innovation of this work is the establishment of a NaOH closed-loop regeneration system. After lithium recovery, the remaining NaOH solution was treated to remove phosphate ions and regenerate the leaching agent by pH adjustment and impurity precipitation, allowing it to be reused in subsequent leaching cycles. The study demonstrated that this regenerated NaOH solution maintained consistent LE over multiple cycles, greatly reducing fresh reagent demand and minimizing wastewater production. An economic analysis further confirmed the process's cost-effectiveness, highlighting that the reuse of NaOH contributes significantly to operational savings. This, along with the simplicity of the process, low-temperature operation, and minimal use of external chemicals, positions the method as a strong candidate for industrial-scale LFP battery recycling. Despite these strengths, several limitations remain. First, the efficiency of NaOH regeneration per cycle and the potential accumulation of byproducts (*e.g.*, unreacted phosphate or Na-based salts) were not quantified. Second, trace impurities (*e.g.*, Al or transition metals) from dismantled battery components may accumulate and affect Fe_3O_4 phase stability or lithium product purity. Third, although phosphate was recovered as Li_3PO_4, the long-term behavior of residual phosphate in the recycled solution was not examined. Over multiple cycles,

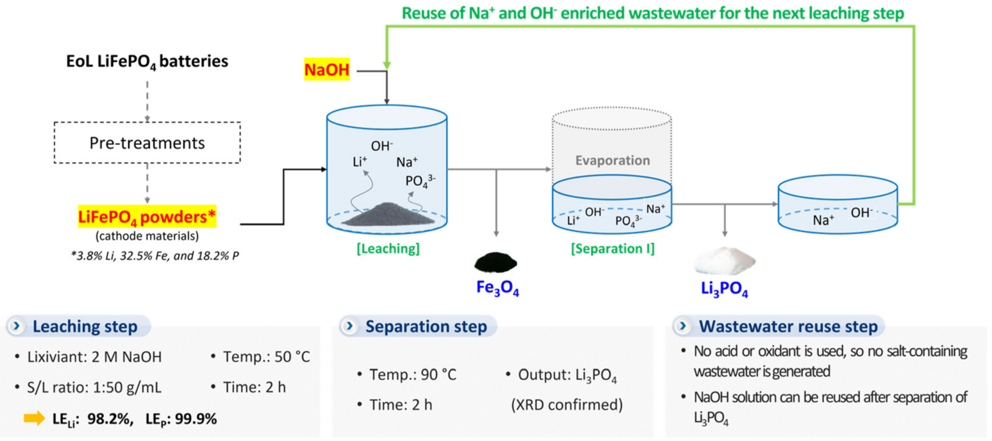


Fig. 9 Conceptual closed-loop hydrometallurgical process for recovery of Fe_3O_4 and Li_3PO_4 from LFP cathode materials using NaOH as a lixiviant. Drawn by the authors based on data reported in ref. 96.

phosphate saturation may reduce product selectivity or crystallization efficiency.

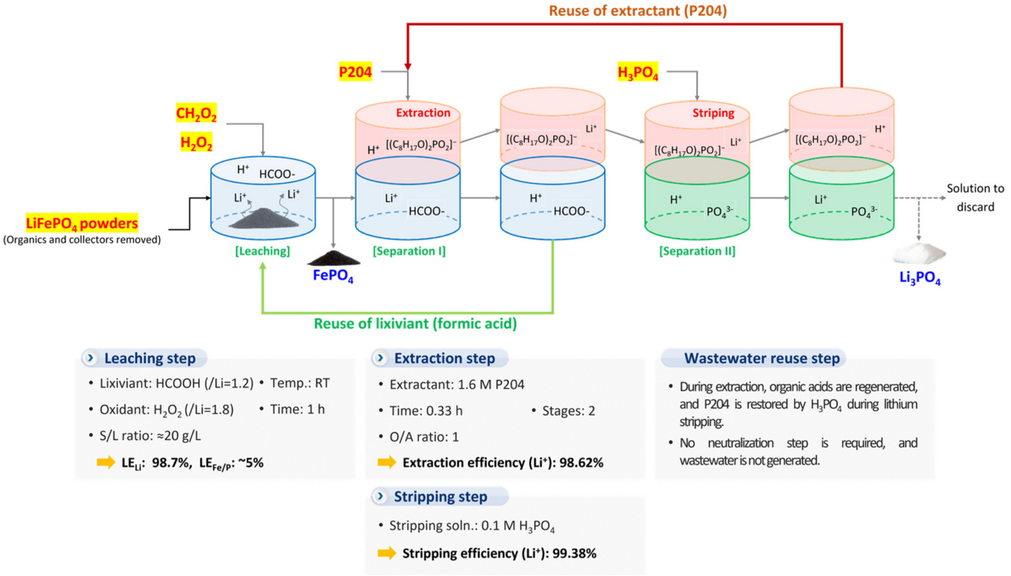
3.5 Organic compound-based lixiviants

3.5.1 Monocarboxylic acids (R-COOH). Representatively, HCOOH serves as an effective and environmentally friendly leaching agent for recovering lithium from EoL LFP due to its strong proton-donating ability, low pK_a (≈3.75), and simple molecular structure. Its short carbon chain and high acidity enable efficient dissolution of lithium under mild conditions. Furthermore, in the presence of hydrogen peroxide, formic acid facilitates selective lithium leaching while minimizing the co-dissolution of iron and phosphorus by stabilizing Fe as solid $FePO_4$, and the general chemical reaction can be represented as follows:

$$2LiFePO_4(s) + 2HCOOH(aq) + H_2O_2(aq) \rightarrow \\ 2Li^+(aq) + 2FePO_4(s) + 2H_2O(l) + 2CO_2(g). \quad (12)$$

The study by Jiang *et al.*[97] explores a highly innovative and sustainable method for recycling EoL LFP batteries, centering on a proton circulation strategy that enables efficient lithium recovery and organic acid regeneration (Fig. 10). The process begins with leaching EoL LFP using monocarboxylic acids (formic, acetic, propanoic, lactic), with H_2O_2 as a green oxidant. Among the tested acids, formic acid exhibited the best performance, achieving 98.7% LE_{Li} with minimal Fe and P dissolution under mild conditions (acid/Li = 1.2, H_2O_2/Li = 1.8, room temperature, and 1 h). After leaching, solvent extraction using P204 enabled lithium separation and simultaneous regeneration of the deprotonated acid. Extraction and stripping efficiencies reached 98.62% and 99.38%, respectively, through two-stage counter-current cycles. The released proton was recycled into the leachate, and dilute H_3PO_4 (0.1 M) selectively stripped lithium over Cu and Al, with separation factors of 148.9 (Li/Cu) and 54.1 (Li/Al). Lithium was ultimately recovered as high-purity Li_3PO_4 (99.5%), and the P204 extractant remained stable over 8 cycles.[97] A key aspect of this study is the implementation of a proton circulation strategy that allows for the regeneration and reuse of the organic acids and extractants, significantly reducing waste production. Although the study did not experimentally validate reuse over multiple leaching cycles, the authors suggest that the regenerated organic acids could be effectively reused, potentially maintaining consistent efficiency and supporting the sustainability of the process. The economic analysis indicated that the process is cost-effective, with a net profit of $0.837 per kilogram of EoL LFP processed. This profitability is largely due to the regeneration and reuse of the expensive organic lixiviant and extractants. While the proton circulation strategy developed in this study demonstrates high lithium recovery efficiency and effective regeneration of leaching and extraction agents, several challenges remain for achieving a fully closed-loop system. The study does not fully evaluate proton balance or buffering capacity over repeated use, key factors for long-term leaching stability. The potential accumulation of trace impurities (*e.g.*, Cu^{2+} and Al^{3+}) and solvent losses due to evaporation or degradation were not quantified. These may impact extraction selectivity, solvent stability, and overall process economics. Addressing these issues will be crucial to realizing a truly circular and viable recycling system.

3.5.2 Deep eutectic solvents (DESs). DESs are gaining prominence in metallurgical processes due to their environmental benefits, tunable properties, and high solubilizing power, making them effective in metal extraction and recycling.[98–102]



Leaching step
- Lixiviant: HCOOH (/Li=1.2) • Temp.: RT
- Oxidant: H_2O_2 (/Li=1.8) • Time: 1 h
- S/L ratio: ≈20 g/L

➡ **LE_{Li}: 98.7%, $LE_{Fe/P}$: ~5%**

Extraction step
- Extractant: 1.6 M P204
- Time: 0.33 h • Stages: 2
- O/A ratio: 1

➡ **Extraction efficiency (Li^+): 98.62%**

Wastewater reuse step
- During extraction, organic acids are regenerated, and P204 is restored by H_3PO_4 during lithium stripping.
- No neutralization step is required, and wastewater is not generated.

Stripping step
- Stripping soln.: 0.1 M H_3PO_4

➡ **Stripping efficiency (Li^+): 99.38%**

Fig. 10 Conceptual closed-loop hydrometallurgical process for recovery of $FePO_4$ from LFP cathodes using CH_2O_2 (formic acid) and H_2O_2 as leaching agents; P204 represents di-(2-ethylhexyl) phosphoric acid. Drawn by the authors based on data reported in ref. 97.

Their low volatility, thermal stability, and potential for electrochemical applications further enhance their appeal. In particular, DESs can be regenerated and reused after hydrometallurgical processes, often through simple recovery methods, such as evaporation or distillation. Their chemical stability and ability to be purified from accumulated contaminants ensure that they maintain efficiency across multiple cycles, contributing to the sustainability and economic viability of metallurgical operations. In typical DES-based leaching systems, protons (H^+) are released from the acidic components of the solvent, contributing to the mildly acidic environment necessary for metal dissolution. These protons facilitate the oxidative leaching of LFP by participating in redox reactions with oxygen, enabling the selective extraction of Li^+ while stabilizing Fe as solid $FePO_4$. This proton-driven mechanism is central to achieving high lithium selectivity in DES-mediated hydrometallurgical processes, and the reaction is shown as follows:

$$4LiFePO_4(s) + 4H^+ + O_2(g) \rightarrow 4Li^+(aq) + 4FePO_4(s) + 2H_2O(l). \tag{13}$$

The study by Zhang *et al.*[103] introduces an innovative and environmentally friendly hydrometallurgical process for the selective recovery of lithium from EoL LFP batteries, using deep eutectic solvents (DESs) as a green alternative to conventional acids (Fig. 11). The process uses chloroacetic acid (CAA) as the hydrogen bond donor and ethanol (EtOH) as the hydrogen bond acceptor in a 1 : 1 molar ratio to form the DES, which serves as the leaching medium. O_2 is used as the oxidant. Under the optimized conditions (70 °C, 6 h, and a solid-to-liquid ratio of 10 g L^{-1}, 0.2 MPa O_2) the process achieved a 100% LE_{Li}, while maintaining iron dissolution below 0.3%, indicating exceptional selectivity. Then, lithium was recovered as $Li_2C_2O_4$ (>90% yield, >99% purity) *via* oxalic acid precipitation at 50 °C, and $FePO_4$ was recovered with 99.7% efficiency from the solid residue. A defining feature of this work is its closed-loop approach, where the DES was reused in at least 3 consecutive leaching cycles without significant performance loss. The study reports that the DES retained its physical structure and leaching ability across 3 cycles. It also highlights that moisture content was the main limiting factor for further reuse and proposes that simple moisture control could extend the number of regeneration cycles even further. The economic analysis conducted in the study supports the feasibility of this approach. By leveraging the low cost of oxygen, the recyclability of DESs, and the high recovery rates of lithium and iron, the process achieved a projected profit of $6032 per ton of EoL LFP ($6.032 per kg LFP) processed.[103] This high profitability underscores the potential for industrial application, especially in regions seeking cleaner, acid-free recycling technologies. However, this economic analysis did not account for the cost of the EoL LFP feedstock, which constitutes a critical input factor. A revised profitability assessment that includes this cost consideration is provided in section 4. Nevertheless, several issues require further attention. Long-term DES stability beyond three cycles was not assessed, nor was a moisture control strategy proposed. The impact of co-leached impurities (*e.g.*, Al and transition metals) and the downstream usability of $Li_2C_2O_4$ for cathode re-synthesis were also not evaluated. In conclusion, this study introduces a novel and green recycling pathway for EoL LFP batteries using a chloroacetic acid–ethanol deep eutectic solvent and oxygen as a clean oxidant. The process achieves near-complete lithium extraction with minimal iron loss and allows for repeated solvent reuse, supporting its economic and environmental viability. However, to realize a truly closed-loop system, future research must address long-term solvent integrity, impurity management, moisture control, and full system lifecycle assessments. These efforts are crucial for scaling this DES-based approach into a fully sustainable industrial process.

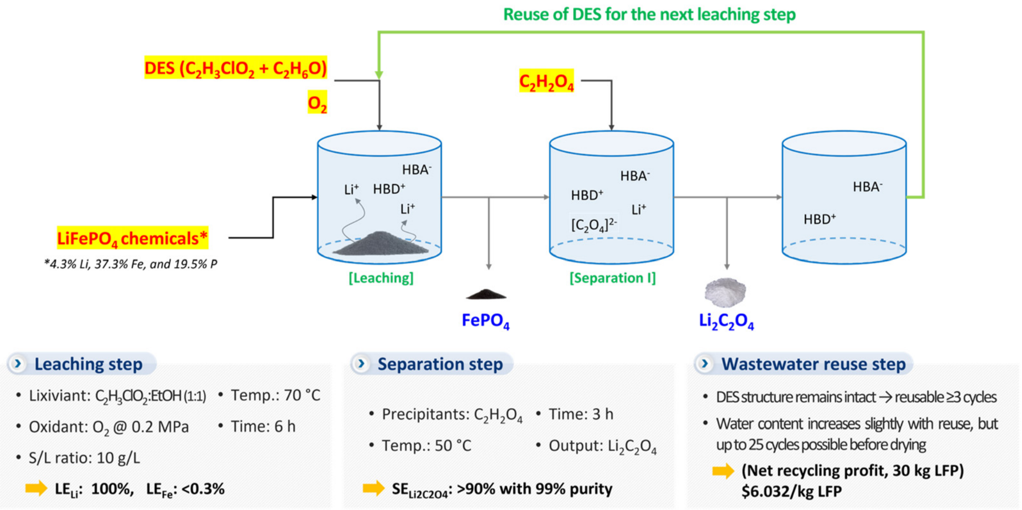


Fig. 11 Conceptual closed-loop hydrometallurgical process for recovery of $FePO_4$ and $Li_2C_2O_4$ from pure LFP chemicals using a DES and O_2 gas as leaching agents; HBA and HBD represent hydrogen bond acceptor and donor, respectively. Drawn by the authors based on data reported in ref. 103.

Table 2 Comparative summary of wastewater reuse strategies across various lixiviant systems in closed-loop hydrometallurgical recycling of LFP batteries

Lixiviant type	Reagents	Wastewater reuse strategy	Reused medium	Recyclability test	Challenges/notes	Ref.
Inorganic acid	H_3PO_4 + H_2O_2	Reacidification after every cycle Lithium recovery every 5 cycles	Mother liquor/ filtrate	5 cycles	The concentration of oxalic acid used was not reported. Accumulation of Na^+ ions was not addressed. The regenerated phosphoric acid process lacks key details such as concentration.	80
	H_3PO_4 + H_2O_2	Mother liquor containing only NaCl which can be reused for the battery discharging step	Mother liquor	NQ[a]	Wastewater was reused only in the discharging step.	69
	H_3PO_4	Evaporate and concentrate to reach the required H_3PO_4 concentration (total volume reduce overtime)	Mother liquor	3 cycles	The viscosity of the leachate build-up after each cycle may inhibit mass transfer and chemical diffusion.	67
Inorganic acidic salt	NaH_2PO_4 + H_2O_2	Reacidified with H_3PO_4 and reused	Post-crystallization brine	NQ	Na^+ accumulation was not monitored. H_3PO_4 was added instead of NaH_2PO_4 from 2 cycles to reacidify. 92% of the initial solution volume was evaporated and reused.	91
	$NaHSO_4$ + H_2O_2	Na_2SO_4 regenerated to $NaHSO_4$ with H_2SO_4 for both recycling leaching or discharging solution	Mother liquor	NQ	Mass balance and impurity accumulation were not evaluated. The sulfuric acid–sodium sulfate reaction is reversible, which may hinder bisulfate ion dissociation. This can reduce LE over multiple wastewater reuse cycles. The Li content was not provided, making it difficult to determine the precise amount of chemicals consumed and needed in subsequent leaching steps.	27
Inorganic oxidizing salt	$Fe_2(SO_4)_3$ + H_2SO_4 + H_2O_2	Full recovery of organic extractant + Fe stripping liquor reused as the lixiviant	Leachate + organic phase	1 cycle	Fe^{3+} losses occurred, as the stripping liquor contained $Fe_2(SO_4)_3$ but its reuse was not addressed. Impurity accumulation in the organic phase was not assessed. A large amount of wastewater was generated during the acid scrubbing and saponification steps.	93
	$Na_2S_2O_8$ + H_2O_2	Na_2SO_4 recovered *via* crystallization and its solution reused as a solvent to prepare the lixiviant	Mother liquor	NQ	The efficiency of $FePO_4$ recovery *via* sieving was not specified, and no quantitative method was provided to assess how much $FePO_4$ detached from the Al foil after leaching.	28
Inorganic alkaline compound	NaOH	Phosphate removed and NaOH regenerated for reuse	NaOH solution/ evaporated solution	NQ	The NaOH solution can be reused indefinitely as it only acts as an oxidizing agent. A critical limitation is that Al foil reacts with NaOH, making this method feasible and environmentally friendly only for treating cathode powders, not whole electrodes.	96
Organic compound	Organic acids + H_2O_2	Proton recycled *via* solvent extraction, organic acid solution, evaporated solution after Li_3PO_4 recovered *via* precipitation	Extractants	NQ	H^+ control is required to regenerate reagents. Only the reuse of organic extractants has been confirmed experimentally.	97
	CAA + EtOH + O_2	DES reused directly after filtration	Same DES medium	5 cycles	Water accumulates in the DES over time, accompanied by a loss of H^+ due to formation of H_2O. pH was not monitored during the process.	103

[a] NQ stands for not quantified.

To summarize, this section highlights the diverse lixiviant systems explored for closed-loop recycling of EoL LFP batteries, with a particular emphasis on wastewater reuse strategies. While these lixiviants differ in their chemical nature and leaching selectivity, a unifying trend among recent studies is the emphasis on wastewater reuse as a strategy to minimize environmental impact and improve process sustainability. Each system employs distinct approaches to reclaim or regenerate wastewater, ranging from direct reuse after filtration or evaporation to full regeneration *via* chemical or electrochemical routes. However, challenges such as impurity accumulation, reusability, reagent stability, and incomplete material balances remain key barriers to achieving truly closed-loop performance. Table 2 provides a comparative overview of the wastewater reuse strategies associated with each lixiviant system discussed in section 3, highlighting the type of reused medium, reuse method, number of validated cycles, and remaining technical limitations.

4. Implications and conclusions

The reviewed studies collectively demonstrate that closed-loop hydrometallurgical processes offer a promising pathway toward sustainable and economically viable recycling of EoL LFP batteries. By focusing on the reuse of aqueous waste streams generated during hydrometallurgical processes, these approaches address one of the major environmental and economical drawbacks of conventional hydrometallurgy, which is wastewater generation, while improving resource efficiency and reducing operating costs. A wide range of lixiviants has been investigated, including inorganic acids (*e.g.*, H_3PO_4 and H_2SO_4), acidic or oxidizing salts ($NaHSO_4$, NaH_2PO_4, $Na_2S_2O_8$, *etc.*), alkaline solutions (NaOH), and green solvents (*e.g.*, organic acids and DESs). These lixiviants were often combined with oxidants such as H_2O_2 or O_2 to enable selective lithium leaching while minimizing co-dissolution of iron and phosphorus. Optimal leaching conditions varied by system but generally achieved over 98% LE_{Li}, with less than 0.05% LE_{Fe} in most cases. Post-leaching separation strategies, such as crystallization, precipitation, and solvent extraction, enabled the efficient recovery of lithium as Li_3PO_4, Li_2CO_3, or $Li_2C_2O_4$ and iron as $FePO_4$ or Fe_2O_3. In several studies, these compounds were shown to be suitable for producing S-LFP. Furthermore, innovative process integrations allowed the regeneration and reuse of wastewater. For example, DES-based wastewater was experimentally verified to be reusable for 5 leaching cycles[103] and phosphate-based wastewater was reacidified for subsequent cycles with minimal performance loss.[80] From an economic perspective, many of the reviewed processes reported net profits ranging from \$0.51 to \$6.03 per kg of EoL LFP batteries (Fig. S4), depending on system design, reagent costs, and product yields.

To further validate these findings, a process-level simulation (Fig. 12) was conducted based on experimentally derived data from the study of Zhang *et al.*[103] and the techno-economic assumptions summarized in Tables S1–S4 of the SI, ensuring a high degree of alignment with real-world conditions such as reagent dosage, product yields (3.10 kg of $Li_2C_2O_4$ and 10.01 kg of $FePO_4$ per process cycle), and equipment utilization. From the second cycle onward, the model assumes 100% reuse of DES-based leachate, reflecting the closed-loop reuse scenario emphasized in this review. Fig. 12a evaluates how country-specific electricity and labor costs affect cumulative net profit over 30 reuse cycles. China and South Korea reached profitability within 4–5 cycles, while regions with higher industrial input costs (*e.g.*, the EU "A" scenario) failed to break even. These trends align with the values shown in Tables S3 and S4, which demonstrate the sensitivity of energy-related costs across regions. To simulate performance decay in reused DESs, a 1% loss in metal recovery per cycle was introduced as shown in Fig. 12b. This leads to a gradual decline in revenue, with peak net profit occurring between the 15th and 20th cycles for cost-efficient countries. The simulation confirms that maintaining leachate stability is critical for extending the economic lifetime of the process. Fig. 12c shows the cumulative net profit calculated under two wastewater loss scenarios (15% and 20%) for China and South Korea. The results indicate that higher loss rates significantly diminish profitability, particularly in South Korea, underscoring the importance of effective wastewater recovery technologies. Given the high viscosity of DESs, filtration can be challenging, making it equally critical to recover as much of the DES as possible to maintain process efficiency and economic viability. Fig. 12d shows the impact of lithium product price fluctuations on economic performance. As lithium-derived revenue is a primary driver of profitability (Table S2), this panel simulates three price reduction scenarios: 40%, 45%, and 50%. A 50% decline in price reduces net profit to near-zero levels across all regions, whereas a 40% decrease still allows for modest profitability in China after approximately 10 reuse cycles. Given that lithium prices have exhibited substantial volatility in recent years due to shifting demand, supply constraints, and policy changes, such sensitivity analyses are essential for evaluating the economic resilience of LFP recycling systems. These results confirm that closed-loop hydrometallurgical processes can not only minimize wastewater treatment cost but also enhance the financial attractiveness of battery recycling operations. However, despite these achievements, none of the studies fully resolve the challenges required for a complete closed-loop system. Several recurring limitations remain across studies:

- Incomplete mass balances

- Most studies do not fully quantify the regeneration efficiency of leaching agents or the amount of fresh reagent needed per cycle.

- Impurity accumulation

- The long-term impact of co-leached metal ions or introduced byproducts (*e.g.*, sodium, phosphate, oxalate, and sulfate ions) on product purity and separation efficiency remains largely unexplored.

- Reusability limits

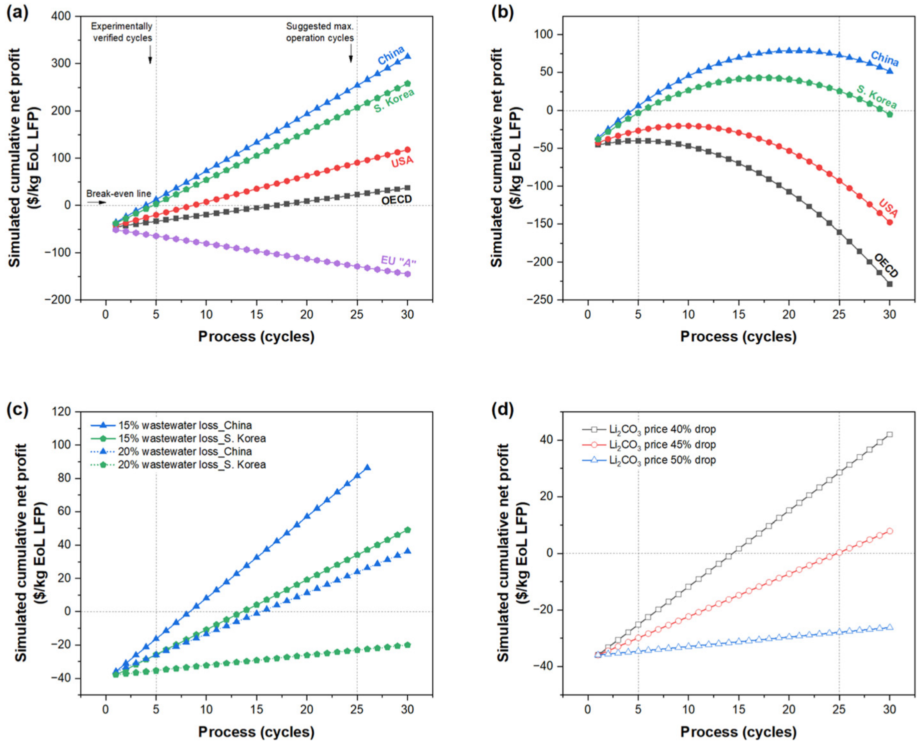


Fig. 12 Simulated process-level net profit for a closed-loop hydrometallurgical process using a DES-based lixiviant for recycling EoL LFP batteries, based on the reported data by Shen *et al.*;[103] (a) effect of country-specific operational costs on net profit per reuse cycle, (b) simulated impact of 1% recovery efficiency of metal compounds decay per cycle; (c) effect of wastewater loss rates on process net profit in China and South Korea, and (d) sensitivity of profit to Li_2CO_3 market price reductions.

- Only a limited number of processes have successfully demonstrated reuse for fewer than 5 cycles, and there are no documented cases of scaling up reuse to industry-relevant cycles (*e.g.*, 10 or more). Furthermore, the impacts of degradation or accumulation over multiple cycles have rarely been quantified.

- Scale-up considerations

- Challenges related to thermal integration, mixing, process control, and impurity management at the industrial scale are often overlooked.

- Lack of life-cycle assessments

- Environmental metrics such as water consumption, energy use, and carbon footprint are omitted in nearly all studies, limiting the ability to assess the environmental benefits of the closed-loop hydrometallurgical process.

To overcome these gaps, future research should prioritize multi-cycle performance testing, closed-loop material and energy tracking, and impurity management strategies. Moreover, integrating life-cycle environmental and economic assessments will be essential to validate these processes for real-world deployment. Finally, extending current frameworks beyond lithium and iron recovery to support full cathode regeneration or cross-chemistry compatibility (*e.g.*, mixed LiBs) would further enhance the circularity and flexibility of closed-loop hydrometallurgical technologies. In conclusion, the adoption of wastewater-reusing, closed-loop hydrometallurgical processes for LFP recycling marks a significant step forward in achieving circularity in battery material recovery. With further refinement, these approaches hold strong promise for industrial-scale implementation aligned with green chemistry and the principles of a circular economy.

Author contributions

Jong-Won Choi: conceptualization, data curation, methodology, and writing – original draft. Hyun-Woo Shim: data curation and validation. Hong-In Kim: funding acquisition and writing – review & editing. Sookyung Kim: funding acquisition and writing – review & editing. Duy Tho Tran: software, validation, and visualization. Mooki Bae: data curation, validation, and writing – original draft.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data for this study are described in the article and SI.

The Supplementary Information includes the Pourbaix diagram of the LFP system, conceptual closed-loop process flow diagrams, and details of the techno-economic assessment. See DOI: **https://doi.org/10.1039/d5gc02987b**.

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