

REVIEW OPEN ACCESS

Advancing Upcycling and Regeneration of Spent LiFePO₄: Failure Mechanisms and Recovery Strategies

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Received: 26 October 2025 | **Revised:** 15 December 2025 | **Accepted:** 16 January 2026

Funding: National Key Research and Development Program of China, Grant/Award Number: 2022YFB2404300; Science and Technology Special Fund of Hainan Province, Grant/Award Number: ZDYF2025GXJS008; Hubei Provincial Science and Technology Innovation Talent Scheme Project, Grant/Award Number: 2024DJA006

Keywords: characterization method | direct regeneration | failure mechanism | LiFePO₄ | spent battery | upcycling

ABSTRACT

With the widespread application of lithium iron phosphate (LFP) batteries in electric vehicles and energy storage fields, the number of retired LFP has increased sharply. Therefore, the development of efficient and environmentally friendly regeneration methods is crucial for the sustainable utilization of resources and environmental protection. In this review, failure mechanism of LFP through advanced characterization technologies is summarized, and upcycling strategies based on the failure mechanism are highlighted. Then, the advantages and challenges of traditional hydrometallurgy, direct regeneration, and upcycling in the recovery of spent LFP (S-LFP) are explored. Among them, upcycling mainly includes strategies such as structural optimization, phase transformation, and conversion into other functional materials. Moreover, the application of materials obtained by upcycling exhibits broaden aspect, including reusing lithium-ion batteries, sodium-ion batteries, and catalytic materials, which significantly improves the economic value of S-LFP cathode materials. Finally, the upcycling method by combining advanced characterization technologies and machine learning to promote the applications and establish a unified economic and environmental analysis system to provide clear analysis standards for relevant research is emphasized.

1 | Introduction

Lithium-ion batteries (LIBs) are widely used in the fields of power and energy storage due to their high energy density, long cycle life, non-toxicity, and stable performance. With the vigorous promotion of the low-carbon concept, the transformation of green travel, and the explosive growth of the energy storage market, it is predicted that the global LIB capacity will increase from 125 to 390 GWh between the year of 2020 and 2030 [1]. In 2026 and 2034, the demand for LIBs will, respectively, reach 139.26 billion dollars and 499.31 billion dollars [2, 3]. The

current mainstream cathode materials for LIBs include lithium iron phosphate (LiFePO₄, LFP), nickel cobalt manganese oxide (NMC), lithium cobalt oxide (LCO), lithium manganese oxide (LMO), and so on [4]. Among them, NMC has good low-temperature performance and high energy density and is mostly used in the high-end EVs field. LCO exhibits a higher voltage platform and is mostly used in the 3C field, such as mobile phones. By comparison, LFP has lower costs due to the absence of precious metals such as Ni and Co. Besides, LFP displays a stable voltage platform and longer cycle life because the

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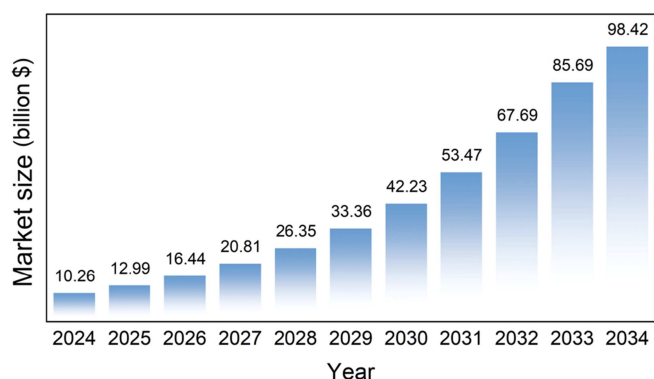


FIGURE 1 | LIB recycling market size and forecast from the year of 2025–2034.

interface reaction by-products do not contain O_2 , which makes it safer. Therefore, it has attracted increasing attention, especially in the field of large-scale energy storage, where its market share is also growing (over 32%) [5]. However, the average lifespan of LIBs is only 5–10 years. In the energy market, when the state of health (SOH) of the battery is below 80% of its initial capacity, it is considered necessary to retire. At this time, retired lithium batteries can be utilized in the energy storage field in a tiered manner. For lithium batteries in the energy storage field, when the SOH is below 60%, they will also be considered scrapped. These lithium batteries are unable to meet the needs of energy storage and power and need to enter the recycling stage [6, 7]. According to statistical data shown in Figure 1, the global LIBs recycling market is expected to reach 98.42 billion dollars in 2034 [8]. It is expected that the recycling of retired LIBs will experience explosive growth in 5–8 years, and the recycling rate of LFP batteries will exceed 58% [9, 10]. Under the background of green economy, the recovery of LFP batteries will become a new hotspot [11].

Common recycling methods for retired LFP include pyrometallurgy, hydrometallurgy, and direct regeneration [12]. Pyrometallurgy requires converting valuable metals into alloys at over 800°C , suffering from high energy consumption, environmental pollution by waste gas or residues, and low Li recovery efficiency [13]. Thus, pyrometallurgy has been seldom used in recent years, and this review does not include this method. By comparison, hydrometallurgy extracts valuable elements via leaching, extraction, and precipitation and re-synthesizes electrode materials, featuring low leaching temperature ($<100^\circ\text{C}$), low energy consumption, and high efficiency, but faces issues like excessive reagent use, long process, acid pollution, and wastewater treatment pressure [14, 15]. Direct regeneration restores cathode structure without destruction, offering short process, low reagent consumption, and high-value regenerated electrodes, effectively avoiding the drawbacks of the other two methods [16, 17]. However, all existing methods only restore material capacity without enhancing electrochemical performance, limiting economic value. Hence, developing new recycling methods for spent LFP (S-LFP) is necessary to meet diverse field demands and expand the application scope.

Upcycling strategies offer a transformative pathway to substantially enhance the economic value of recovered products from S-LFP, thereby improving the profitability of recycling

processes and fostering the sustainable development of the battery-recycling sector [18]. Since the market price of regenerated LFP (R-LFP) is primarily determined by its electrochemical performance, such as capacity retention, rate capability, and cycle stability, value enhancement can be directly achieved through targeted modification of the LFP lattice or its surface carbon layer via doping or coating protocols [19]. Element doping optimizes electronic conductivity, while carbon coating mitigates particle agglomeration and enhances interfacial stability. Meanwhile, the partial replacement of Fe with Mn in the olivine lattice represents a straightforward and effective approach to raise the working potential and transforms into a marked increase in energy density that significantly broadens the application space of the upcycled cathode, particularly in high-energy-density LIBs [20]. Beyond LIBs, the product portfolio can be diversified to encompass post-lithium energy storage systems (such as sodium-ion or potassium-ion battery electrodes) and functional materials including photocatalysts and oxygen evolution reaction (OER) catalysts, thereby accessing wider markets with higher value and/or functionality than the original materials [21, 22]. Nevertheless, S-LFP feedstocks inherently present challenges related to SOH, failure modes (such as structural degradation and electrolyte decomposition), and impurity inventory from current collectors or electrolytes [23]. Such heterogeneity can lead to inconsistent reaction kinetics, compromised product quality, and increased process complexity. Therefore, it is essential to carefully select appropriate recycling routes to balance technical feasibility, process cost, and economic rationality, ensuring the scalability and competitiveness of upcycling technologies.

Tracing the failure mechanism of S-LFP is a prerequisite for rationally selecting recycling protocols. Pretreating S-LFP based on failure mechanisms yields a large quantity of feedstocks with highly consistent compositions and structures, which can be rapidly matched to the optimal recycling strategies. Such pretreatment shortens the production process and elevates overall process efficiency. More fundamentally, elucidating failure mechanisms can provide precise design targets for upcycling technologies. By specifically strengthening the components most prone to failure, the performance of R-LFP can be enhanced. For instance, the coated carbon layer is susceptible to cracking, and upcycling technologies can improve the stability of this coated carbon layer. Additionally, to address the issue of the relatively low energy density of LFP, upcycling technologies can resolve this problem by increasing the operating voltage of LFP. Consequently, clarifying failure mechanisms is not only of great significance but also an indispensable means to achieve the closed-loop valorization of S-LFP.

The review starts with a detailed exposition of failure mechanisms of LFP and respective characterization techniques, including loss of active-lithium, FePO_4 (FP) parasitic-phase formation, crystallite fracture, and disruption of the solid electrolyte interphase (SEI) film. Advanced characterization techniques can show the failure pathway explicitly. Subsequently, conventional hydrometallurgical and direct regeneration are analyzed through economic-environmental standards. The advantages and challenges of high energy consumption, heavy wastewater generation, and feedstock specificity are clarified. The discussion then focuses on three upcycling paradigms that systematically improve the economic value of regenerated

products: (1) structural optimization to yield high-performance LFP, (2) phase engineering toward lithium iron manganese phosphate (LFMP) with elevated working potentials, and (3) transformation of S-LFP into post-lithium-ion battery electrodes and heterogeneous catalytic architectures. By expanding high-value recycles, these strategies extend the utilization horizon of S-LFP. The objective of this article is to furnish a comprehensive theoretical framework and scientific substantiation for the large-scale deployment of LFP battery upcycling technology.

2 | Failure Mechanisms and Failure Analysis of LFP

2.1 | Failure Mechanism of LFP

Studying the methods of recycling and regenerating retired batteries is an important issue in reducing environmental pollution and achieving resource recycling. In this article, battery recycling refers to the extraction of valuable metal elements from batteries through physical or chemical processes. Battery regeneration refers to the chemical repair of failed LFP cathode materials, allowing them to be reused in new batteries or converted into other high-performance materials without the need to convert LFP into iron salts or oxides during this process.

Clarifying the failure mechanism of LFP is the primary prerequisite for studying green and efficient regeneration technology. LFP has a stable olivine-type crystal structure (space group *Pnma*), belonging to the orthorhombic crystal system (also known as the rhombohedral system, $a \neq b \neq c$), and the lattice structure of LFP is shown in Figure 2A. Its olivine-type structure consists of FeO_6 octahedra, LiO_6 octahedra, and PO_4 tetrahedra [28]. FeO_6 octahedra, LiO_6 octahedra, and PO_4 tetrahedra alternate along the *a*-axis direction to form a layered structure. One PO_4 tetrahedron shares a common edge with one FeO_6 octahedra and two LiO_6 octahedra along the *c*-axis direction, forming a stable 3D spatial network structure [29, 30]. The P-O bond with high bonding energy in LFP connects FeO_6 octahedra, forming a framework structure that contributes to the high dynamic and thermodynamic stability of its crystal skeleton [31]. PO_4 tetrahedra connect FeO_6 octahedra that are parallel to each other, so Li^+ can only diffuse in one dimension along the *b*-axis direction of the orthorhombic crystal system. The adjacent FeO_6 octahedra limit Li migration, so an irreversible phase transition may occur inside LFP after long-term cycling [32, 33].

Inside the LFP particles, lithium ions shuttle repeatedly between LFP and graphite. When lithium ions migrate into graphite, lithium vacancies will form in LFP. Fe ions will diffuse into these vacancies under the action of potential difference, while Li ions returning from graphite will occupy the vacant Fe sites, which is known as Li-Fe anti-site defects [34–36]. This phenomenon significantly reduces the Li^+ diffusion coefficient. Some of the lithium that migrates to graphite is confined to graphite and cannot return to LFP, forming “dead lithium.” Another part participates in the construction of the SEI film, which results in the loss of active lithium in LFP, directly leading to a decrease in battery capacity [27, 37, 38]. Lithium deficiency causes a charge imbalance in the LFP crystal phase. In order to balance the charge, Fe^{2+} is oxidized to Fe^{3+} , forming the FP phase [39–41]. The FP phase, formed by Fe^{3+} , covers the LFP phase and induces the formation of disordered regions between the LFP phase and the FP phase [42, 43].

At the particle level, under long-term cycling, the conductive carbon layer on the surface of LFP particles is damaged, and the particles as a whole may form cracks or even break [44]. Padhi et al. [45] found that this phenomenon is caused by the repeated de-intercalation of Li^+ , which transforms the single-phase LFP structure inside the particles into a sharp interface LFP/FP two-phase structure, resulting in internal strain. Through calculations, Morgan et al. [46] found that during the detachment of lithium ions, the particle volume shrinks by 5.03% along the *a*-axis and elongates by 1.92% along the *c*-axis. The stress generated by volume expansion may be applied to the conductive carbon layer at the local interface or surface, and this uneven change can cause cracking and fragmentation of the crystal surface and the conductive carbon layer covering the particle surface [47], inducing high polarization of the battery, which is also one of the reasons for capacity degradation [48]. The damage to the conductive carbon layer will reduce the electronic conductivity of the battery, affecting the transport of ions and electrons. The formation of cracks will increase the specific surface area of the material, reduce particle size, and intensify the occurrence of side reactions between LFP and electrolyte [49]. New side reactions will thicken the SEI film, and continuous thickening will further reduce battery capacity and cycling performance [42].

At the interface between LFP particles and electrolyte, during the first charge–discharge process, the electrode material reacts with the electrolyte at the solid–liquid interface to form a passive layer covering the LFP, known as the SEI film [50–52]. The SEI film initially plays a role in preventing electron transfer and preventing further degradation of the electrolyte. But as the cycle progresses, the SEI film is repeatedly reconstructed, and the unstable SEI film disappears. In some areas, the SEI film thickens [53]. In addition, the inorganic components LiF and Li_2CO_3 in the SEI film decrease, the organic component fluorophosphate increases, and the electrolyte desolvation process weakens, resulting in co-doping of solvent molecules in LFP. Increasing interface impedance hinders Li^+ transport [42]. When the SEI film of the anode thickens, it not only increases the impedance of the battery but also leads to the failure of lithium ions to timely embed into the layered structure of the anode material after obtaining electrons on the surface of the anode. Instead, they deposit on the surface in the form of metallic lithium to form lithium dendrites, resulting in lithium precipitation phenomenon [54–56]. The formation of lithium dendrites not only consumes a large amount of active lithium but also may puncture the separator, causing internal short circuits in the battery [57], posing a great threat to battery safety.

In summary, there are four main reasons for the failure of LIBs. As shown in Figure 2B–D, Issue 1: the loss of active lithium during charging and discharging processes, Issue 2: the formation of Fe^{3+} , Issue 3: the inevitable particle breakage under long-term cycling, and Issue 4: the damage of conductive carbon layer. The schematic diagram of LFP failure mechanism is shown in Figure 2E. In light of the issues above, the output methods of hydrometallurgy for S-LFP involve leaching of Li, Fe, and P, and then recovering them through separate precipitation, or selective leaching of one element and recovery [58]. The direct regeneration method focuses on the supplementation of active lithium, reduction of Fe^{3+} , and repair of the crystal structure and conductive carbon layer in spent battery materials, thereby achieving the recovery of electrochemical performance.

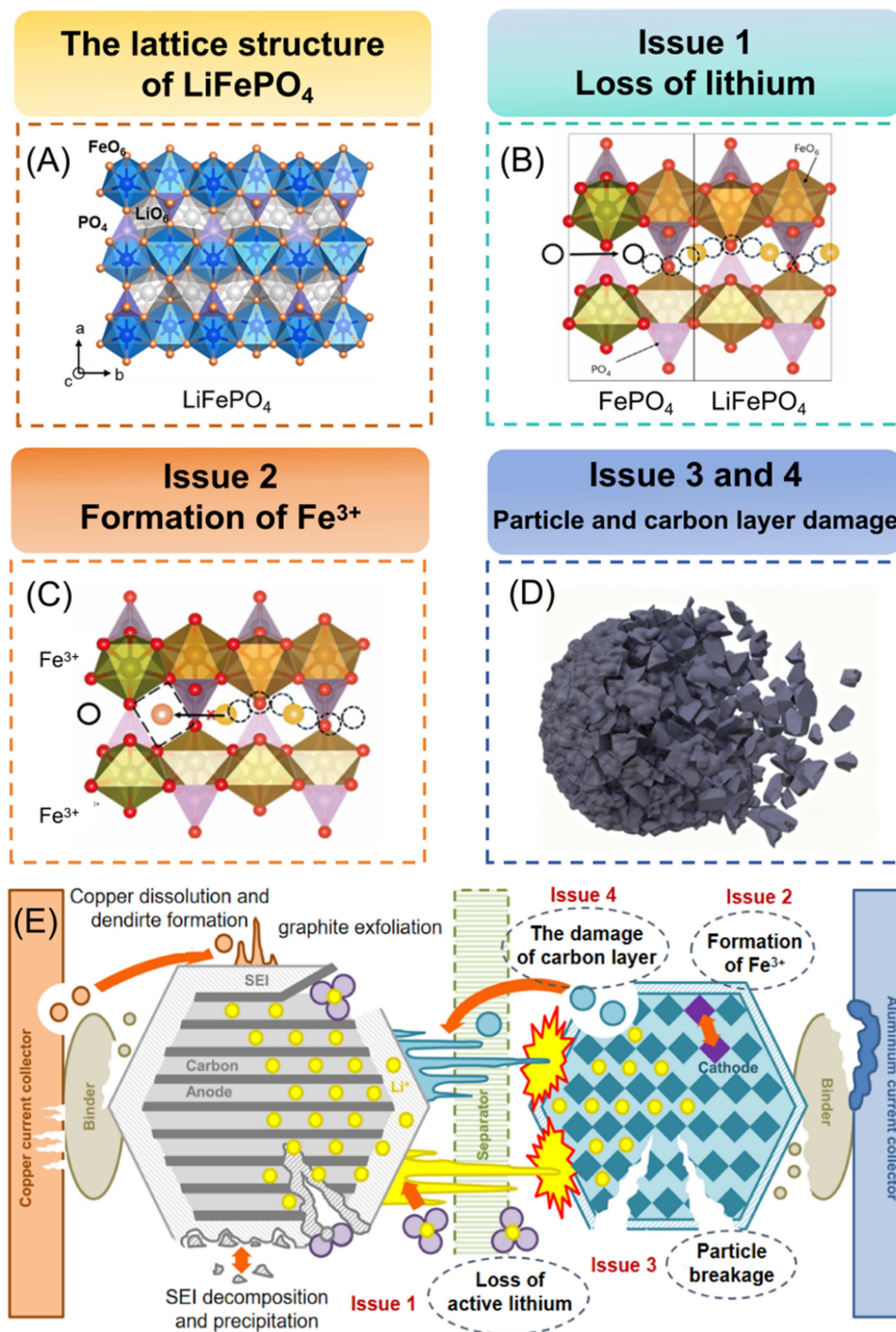


FIGURE 2 | (A) The lattice structure of LFP. Reproduced with permission: Copyright 2023, American Chemical Society [24]. Schematic diagram of (B) Issue 1: loss of lithium. Reproduced with permission: Copyright 2025, Springer Nature [25]. (C) Issue 2: formation of Fe^{3+} . Reproduced with permission: Copyright 2025, Springer Nature [25]. (D) Issues 3 and 4: particle and carbon layer damage. Reproduced with permission: Copyright 2023, Wiley [26]. (E) LFP failure mechanism. Reproduced with permission: Copyright 2017, Elsevier [27].

2.2 | Characterization Methods for LFP Failure Analysis

The adoption of appropriate characterization methods is the basis for understanding the failure mechanism of batteries. By characterizing the physical properties and electrochemical measurements of failed batteries, the root cause of failure can be accurately located. Common methods for characterizing

physical properties include morphology characterization, structural characterization, interface characterization, valence state detection, and element/component determination [59]. In order to obtain more accurate and authentic physical property information, some advanced physical property characterization techniques have also been widely applied in battery failure analysis research, including in situ characterization techniques, multi-combination characterization techniques, and so on.

The main reason for the failure of S-LFP is the loss of active lithium [60]. To measure the Li content of S-LFP, Tang et al. [61] compared the Li/Fe ratio of three different samples using inductively coupled plasma-optical emission spectroscopy (ICP-OES) and found significant lithium loss in S-LFP, with a Li/Fe ratio of only 0.88, lower than the theoretical value of 1.0 (Figure 3A). In situ optical microscopy (OM) can simulate the real working environment of batteries and observe macroscopic changes in electrode structure. Hogrefe et al. [62] characterized the lithium deposition growth and dissolution of graphite/LFP full cells during 1 C charging through this way (Figure 3B) and found that uneven lithium deposition appeared in a tree-like structure after 25 min of constant current charging. Subsequently, constant voltage charging was performed, and the lithium deposition continued to thicken, reaching its maximum thickness at 39 min before starting to dissolve. At 62 min, “dead lithium” residue can be seen, and the lithium metal reacts with the electrolyte to form an SEI, which reduces the electrochemical performance of the battery. The other reason for the failure of S-LFP is the formation of Fe^{3+} . Tang et al. [61] also analyzed the valence state changes and composition of S-LFP,

and they found that Li deficiency induces the formation of FP phase and Fe^{3+} , as evidenced by X-ray photoelectron spectroscopy (XPS) (Figure 3C) and X-ray diffraction (XRD). There are both Fe^{2+} and Fe^{3+} peaks in XPS spectra, with Fe^{2+} peak at 723.8 eV and 710 eV and Fe^{3+} peak at 726.4 eV and 711.1 eV. XRD spectra show diffraction peaks attributed to FP at 16° and 18.5° , indicating that S-LFP is in a state of coexistence of LFP and FP. The lithium vacancies caused by lithium deficiency can also lead to Li-Fe anti-site defects. Lin et al. [63] used XRD Rietveld refinement (Figure 3D) to verify that S-LFP indeed has 19.86% FP phase and 6.84% Li-Fe anti-site defects. Fourier transform infrared (FTIR) is also a commonly used method for detecting Li-Fe anti-site defects. The FTIR results of Jia et al. [64] showed that the Li-Fe anti-site peak of S-LFP is at 996.2 cm^{-1} , which is higher than that of repaired LFP (RSLFP@NC). The lower wavenumber (1021.2 cm^{-1}) indicates the presence of a large number of anti-site defects in S-LFP [65]. In situ XRD can observe the phase transition process in the S-LFP heat treatment reaction without damaging the material (Figure 3E). At a low temperature, LFP and FP phases coexist, and the characteristic peak of the FP phase gradually disappears

Characterization Methods for LFP Failure Analysis

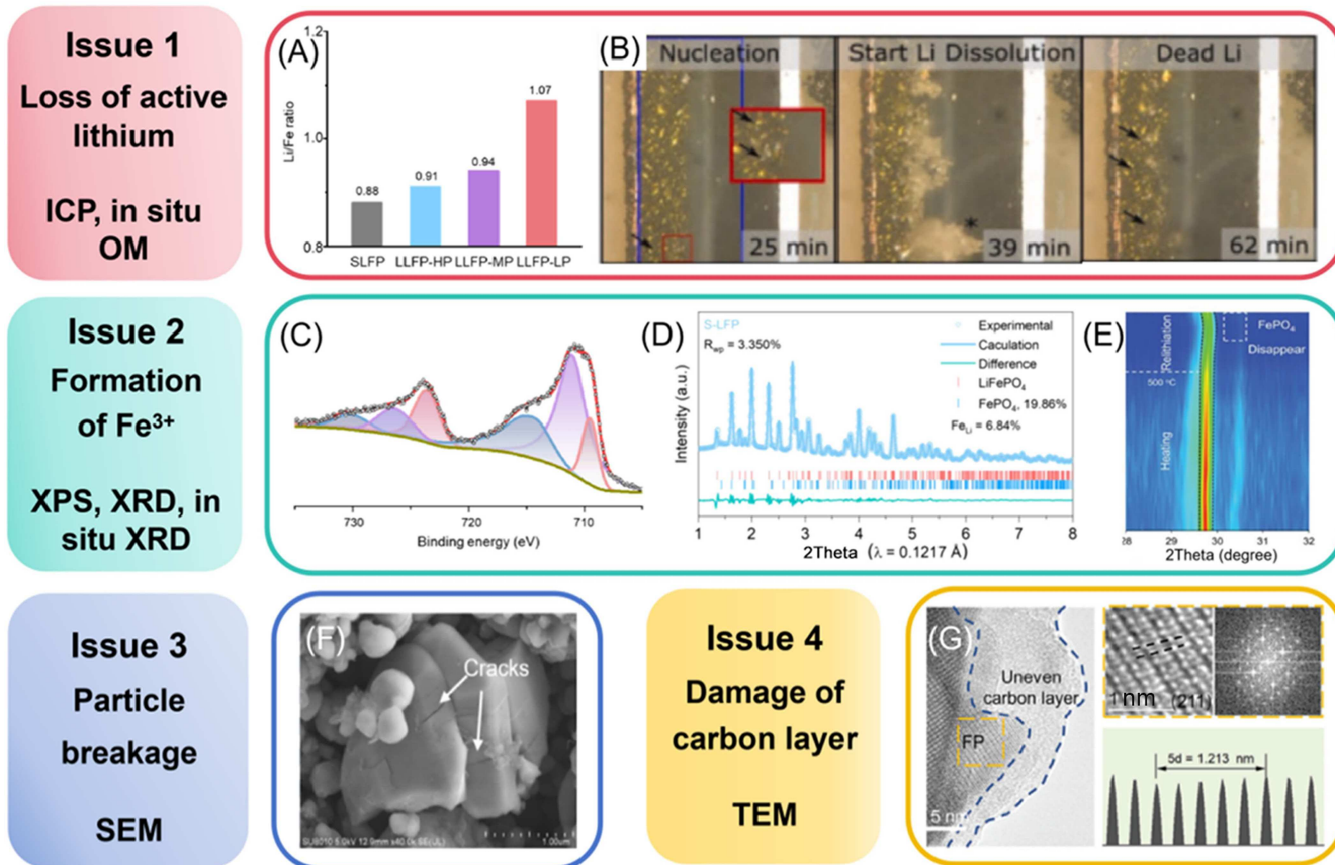


FIGURE 3 | (A) ICP of S-LFP and other samples. Reproduced with permission: Copyright 2025, Wiley [61]. (B) In situ OM of a full cell during a charging process. Reproduced with permission: Copyright 2023, Elsevier [62]. (C) Fe 2p XPS spectrum of S-LFP. Reproduced with permission: Copyright 2025, Wiley [61]. (D) XRD pattern and Rietveld refinement of S-LFP. Reproduced with permission: Copyright 2025, Chinese Chemical Society [63]. (E) Contour map of in situ XRD during heat treatment. Reproduced with permission: Copyright 2023, Wiley [64]. (F) SEM and (G) TEM images of S-LFP. Reproduced with permission: Copyright 2025, Wiley [61].

after reaching 500°C, indicating that the FP phase is transformed into the LFP phase after appropriate lithiation, further proving the existence of LFP and FP phases inside S-LFP. The FP phase and Li-Fe anti-site defects generated by Li loss will hinder the efficiency of Li^+ transport, and for the repair and regeneration of S-LFP, special attention needs to be paid to the supplementation of active lithium.

Another main reason for LFP failure is particle breakage. To observe the surface morphology and structural defects of particles, Tang et al. [61] conducted research using scanning electron microscopy (SEM) (Figure 3F) and transmission electron microscopy (TEM) (Figure 3G). The SEM image shows that under the influence of long-term stress accumulation, there are obvious cracks on the surface of S-LFP particles. TEM confirmed the presence of an uneven carbon coating on the surface of the particles, indicating damage to the carbon layer and SEI film on the surface. In addition, lattice stripes with a spacing of 2.426 Å were discovered, corresponding to the FP (211) crystal plane, further indicating the generation of impurity phases. To study the chemical phase distribution of micrometer-level single crystals, Boesenberg et al. [66] combined scanning transmission X-ray microscopy (STXM) with full-field transmission X-ray microscopy (FF TXM) and X-ray near-edge spectroscopy (XANES) to analyze the phase transition mechanism and microstructure. The areas with reduced brightness contrast in the FF TXM image correspond to regions with lower mass density, and there are cracks or voids inside the surface particles. The diagram obtained by linear combination fitting of XANES data shows that there is crystal superposition in some areas, indicating that some Fe^{2+} in the delithiated crystal is oxidized to Fe^{3+} , meaning that the delithiated crystal is composed of LFP phase and FP phase superposition, and there are also some areas with low brightness contrast. It can be inferred that the cracks on the surface and inside of the particles are caused by the tensile stress generated by the lattice mismatch between the LFP phase and the FP phase, which leads to the cracks cracking along the c-axis [67]. Particle breakage or surface cracks can reduce the electronic conductivity of the material, and the carbon coating can also rupture, reducing the electrochemical performance of the battery.

This article mainly focuses on several representative physical characterization methods that are frequently used in the failure analysis of LFP and introduces the applications of various methods. By accurately diagnosing and analyzing the process of battery failure, a fundamental understanding of the failure mechanism can contribute to the progress of battery regeneration research and promote green and sustainable energy development.

3 | General Methods for Regeneration of S-LFP

3.1 | Hydrometallurgy

Hydrometallurgy utilizes chemical reagents to fully or selectively leach valuable metals from cathode materials, and the recovered products are mostly metals or metal oxides [68–70]. Hydrometallurgy is mainly divided into three categories: acid leaching, acid-free leaching, and deep eutectic solvents (DESs) leaching [71–73]. Before commonly used leaching, it is not only necessary to dismantle, screen, and separate the collector from

spent batteries but also to pretreat the materials, including the removal of conductive carbon and binders and the harmless treatment of the electrolyte. In addition, various auxiliary methods are often used to accelerate the leaching process, including mechanochemistry, ultrasound, microwave heating, Joule heating, solvothermal, and supercritical liquids. Compared with pyrometallurgy, hydrometallurgy has lower energy consumption and greenhouse gas emissions, but the separation and purification steps are more complex, and the acidic wastewater generated also requires additional treatment [74].

3.1.1 | Acid Leaching

Acid leaching includes inorganic acid leaching, organic acid leaching, and inorganic/organic acid composite leaching. Inorganic acids have low cost and fast leaching rate and can dissolve almost all metals [75, 76]. Cong et al. [77] used a combination of $\text{H}_2\text{SO}_4\text{--H}_2\text{O}_2$ to synergistically leach Li and Fe from S-LFP. Under the optimal reaction conditions, the leaching efficiency of Li reached 95.23%, while Fe and P were retained in the residue. In addition to H_2SO_4 , H_3PO_4 and HCl are also commonly used in the acid leaching method. Based on the redox interaction between NMC and LFP, Zhou et al. [78] proposed a one-step separation method, which enables the recovery of Fe without additional oxidants. As S-LFP dissolves in H_3PO_4 , Fe^{2+} undergoes an oxidation reaction with NMC to form Fe^{3+} , generating $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$. Meanwhile, the high-valent Li, Mn, Ni, and Co are reduced to low-valent states, with their leaching efficiencies reaching 99.6%, 99.1%, 98.9%, and 97.3%, respectively, under this condition (Figure 4A). All these results demonstrate the feasibility of recovering LFP using inorganic acids. Li et al. [82] first used HCl solution to leach Fe ions from LFP. Since no oxidant was added, most of the iron ions existed in the form of Fe^{2+} at this stage. Subsequently, an ionic liquid-based aqueous biphasic system (IL-ABS) was employed to separate lithium and iron from the filtrate. The IL-ABS consists of $[\text{P44414}]\text{Cl} \cdot \text{NaCl}$. By adopting a two-end cross-flow solvent extraction process, 98% of iron can be leached, while Li^+ remains in the aqueous phase. Adding H_2O can precipitate 97% of iron ions, obtaining $\text{Fe}(\text{OH})_2$ and $\text{Fe}(\text{OH})_3$ with a purity of nearly 100%. Adding K_3PO_4 solution can precipitate 90% of Li, yielding Li_3PO_4 with a purity of 99.5%. When this method is used to recover LFP, the extraction rate of Fe ions can still reach 90% after the regenerated IL is reused four times. It can be found that the leaching efficiency of inorganic acids is very high, but inorganic acids are prone to corrosion of instruments and equipment, and the treatment of wastewater and toxic elements generated is also a major challenge [58].

For organic acids, their characteristics of being green, mild, easily degradable, and recyclable have made them favored by many researchers [83]. Integrating the principles of green chemistry, Yang et al. [84] used a small amount of CH_3COOH to leach lithium from S-LFP and oxidized Fe^{2+} to Fe^{3+} with H_2O_2 . Finally, they successfully recovered lithium and iron in the forms of Li_2CO_3 and FePO_4 . The leaching efficiency of Li^+ could reach 94.08% in this work. However, CH_3COOH has weak acidity, resulting in an unsatisfactory leaching effect. Zeng et al. [85] selected three different organic acids: citric acid, oxalic acid, and lactic acid, combining them with H_2O_2 to form a leaching system for the S-LFP cathode material. The

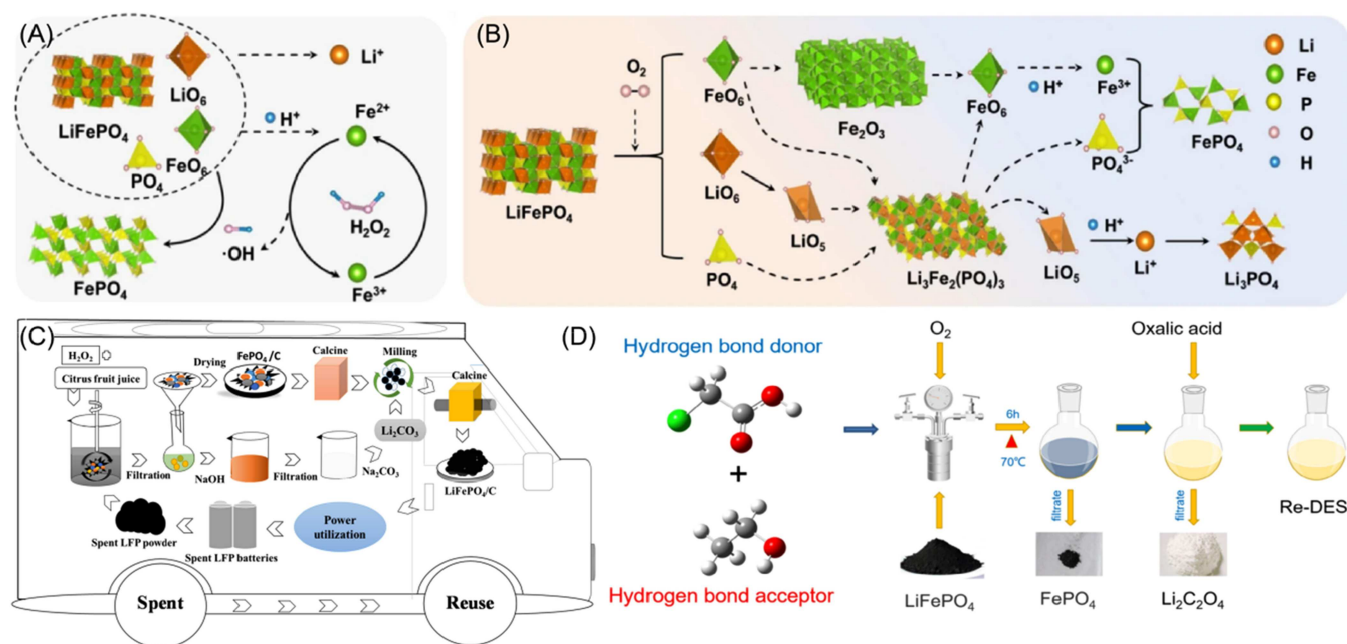


FIGURE 4 | The mechanism and flowchart of hydrometallurgy recovery: (A) Leaching S-LFP using inorganic acid composed of H_3PO_4 . Reproduced with permission: Copyright 2023, Elsevier [78]. (B) Leaching S-LFP using organic acid composed of lemon juice. Reproduced with permission: Copyright 2020, Elsevier [79]. (C) $\text{H}_2\text{SO}_4\text{--H}_2\text{O}_2$ and (D) O_2 roasting and acid leaching reaction mechanism. Reproduced with permission: Copyright 2025, Nonferrous Metals Society of China [80]. (D) Leaching S-LFP using DES composed of chloroacetic acid and ethanol. Reproduced with permission: Copyright 2025, Elsevier [81].

study found that lactic acid has a lower coordination ability with Fe^{3+} , which means it is easier to achieve the separation of iron and lithium. Under the optimal experimental conditions, lactic acid can selectively leach more than 99% of lithium and less than 2% of iron. In addition to these organic compounds, citrus juices rich in citric acid and malic acid can also be used as leaching agents. Kumar et al. [79] discovered that among lemon, apple, and orange juices, lemon juice exhibits the best leaching performance. The flow diagram of the leaching process using lemon juice is shown in Figure 4B. After adding H_2O_2 and reacting at room temperature, the leaching efficiencies of Li and Fe reach 94.83% and 4.05%, respectively. Li and Fe are recovered in the forms of Li_2CO_3 and FePO_4 , which are further used to re-prepare new LFP cathode materials. Meanwhile, other elements such as Cu, Al, and P can also be leached out. The newly prepared LFP cathode material has a discharge capacity of 155.3 mAh g^{-1} at 0.1 C and a capacity retention rate of 98.3% after 100 cycles at 1 C. Most commonly used organic acids are biomass-based acids, which have a negligible impact on the environment and conform to the concept of green chemistry. However, their industrial application is difficult to promote due to the limitation of recovery costs.

3.1.2 | Acid-Free Leaching

Typical acid-free leaching methods include mechanochemically assisted water leaching, oxidant leaching, and alkaline leaching. Mechanochemistry aims to use externally applied mechanical force to destroy material structures, accelerate the leaching of Li^+ , and reduce the usage of organic or inorganic acids [86]. Alternatively, it directly employs water, inorganic salts, or organic salts for leaching [87, 88]. To avoid the use of acid,

Zhang et al. [89] mixed S-LFP powder with sodium citrate and H_2O_2 and ball-milled the mixture, achieving leaching efficiencies of 98.9% for Li and 5.1% for Fe. Subsequently, Na_2CO_3 was added to precipitate Li in the form of Li_2CO_3 . This method uses non-toxic and renewable reagents, thus exhibiting excellent environmental friendliness. Besides, the recovery of mixed cathode materials via mechanochemical methods is also a major research focus at present. Jiang et al. [90] mixed LFP and LCO and ball-milled the mixture to destroy the crystal structure of the mixed materials. This process promoted the oxidation of Fe^{2+} and the reduction of Co^{3+} . With the addition of a small amount of H_2SO_4 , leaching efficiencies of 99.9% for Li and 88.6% for Co from different LIBs were achieved.

Leaching with oxidants can avoid environmental pollution and resource waste, and the selection of a suitable oxidant is crucial. To achieve a selective leaching system of S-LFP and NMC, Hua et al. [91] used water-soluble $\text{NH}_4\text{Fe}(\text{SO}_4)_2$ as an oxidant-reductant to synergistically recover valuable metals from spent NMC and LFP. H_2O was added to the NMC/LFP/ $\text{NH}_4\text{Fe}(\text{SO}_4)_2$ mixture, and the reaction was carried out. The residue obtained by filtration was mixed with an $\text{H}_3\text{PO}_4/\text{NH}_4\text{H}_2\text{PO}_4$ buffer solution to regenerate $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$. Under this method, the leaching efficiencies of Li, Mn, Co, and Ni were 97.8%, 98.3%, 98.9%, and 99.0%, respectively. This study proves that the introduction of iron ions can effectively extract metal ions. Based on the unique acidity and oxidability of sodium phosphomolybdate (PMS) with the Keggin-type structure $(\text{PMo}_{12}\text{O}_{40})^{3-}$, Ren et al. [92] used it as an oxidant for the selective extraction of lithium from S-LFP. The reduced PMS can be re-oxidized and reused by activating O_2 into superoxide anions (O_2^-), and the lithium extraction efficiency reaches as high as 96.79%. Moreover, PMS can be recycled to reduce recovery costs. After being re-oxidized and reused 10 times, the

lithium extraction efficiency of PMS remains over 91%. Additionally, Du et al. [93] proposed an alkaline pressure leaching process with the synergistic effect of NaOH and $\text{Ca}(\text{OH})_2$, where NaOH serves as the leaching agent and $\text{Ca}(\text{OH})_2$ serves as the additive. The two substances are mixed and placed in a sealed reactor, and products containing 0.3% P, 0.02% Fe, and 97.88% Li can be obtained after reaction. Compared with acid leaching, alkaline leaching has stronger selectivity for different metal ions, enabling it to preferentially dissolve target metals and leave impurities such as iron and copper in the residue for easier subsequent separation.

H_2O_2 acts as an oxidant in most leaching strategies. This is because H_2O_2 is not only low in cost and strong in oxidizing capacity but also produces only O_2 and H_2O as by-products when used as an oxidant. These by-products do not cause environmental pollution, which conforms to the concept of green chemistry [94]. According to a report by Qiu et al. [95], H_2O_2 was used to leach S-LFP cathode materials, and in this case, H_2O_2 served as both an oxidant and a leaching agent. After leaching, 97.6% of Li could be leached, and FePO_4 powder was obtained simultaneously. Adding Na_2CO_3 to precipitate Li^+ yielded Li_2CO_3 . Subsequently, the two substances (Li_2CO_3 and FePO_4 powder) were mixed, with citric acid added as a reducing agent, followed by appropriate annealing treatment. The R-LFP had a capacity of 144 mAh g^{-1} at 1 C, and the capacity loss was less than 1% after 100 cycles, exhibiting excellent rate performance and cycling stability. In addition to using reagents such as H_2O_2 as oxidants, oxidation under air has also attracted the attention of researchers. Wang et al. [80] compared the leaching efficiencies of S-LFP with H_2SO_4 after oxidation under air and H_2O_2 conditions, respectively. They found that after changing the experimental conditions, although the air oxidation- H_2SO_4 method avoided the use of H_2O_2 , featuring low emissions and simpler, safer operation, Fe was inevitably leached out, which reduced the leaching selectivity and increased the consumption of H_2SO_4 . This phenomenon is attributed to the different leaching mechanisms of the two systems: the diagrams of the solid-liquid-solid conversion mechanism for air oxidation- H_2SO_4 and the solid-solid conversion mechanism for H_2O_2 - H_2SO_4 are shown in Figure 4C,D, respectively. Therefore, selecting an appropriate oxidation method is also crucial for the recovery of S-LFP.

3.1.3 | DESs Leaching

Although the aforementioned methods can achieve efficient leaching of metal ions, they are all inseparable from the massive consumption of chemical reagents and the problem of acid pollution. Therefore, exploring environmentally friendly and low-cost leaching methods is also a major research direction in hydrometallurgy. In recent years, researchers have found that the strong intermolecular interactions of DESs can dissolve the cathode materials of LIBs and efficiently leach the metal elements therein [96, 97]. DES is composed of hydrogen bond donor (HBD) and hydrogen bond acceptor (HBA) mixed in a certain ratio, with a melting point significantly lower than that of its individual components and the ability to be recycled [98]. Based on the acidity of DES and the oxidizing property of oxygen, Zhang et al. [81] mixed chloroacetic acid and ethanol to form DES, and oxygen was used to oxidize Fe^{2+} to Fe^{3+} (Figure 4E). Through this method, the leaching rate of Li is

close to 100%, and 99.7% of FePO_4 can be recovered by filtration. Moreover, the leaching efficiency of the reused DES does not decrease either. However, binary DESs generally have excessively high viscosity, which impairs leaching efficiency. To address this issue, Zou et al. [99] compared ternary DES systems with different compositions and found that the DES composed of choline chloride, ethylene glycol, and maleic acid exhibited the highest leaching efficiency for spent LIBs. After adding the black mass of spent LIBs to this ternary DES, the leaching of over 99% of Ni, Co, Mn, Li, and Fe was observed. Then NaOH was added to adjust the pH of the leachate, resulting in the precipitation of $\text{Fe}(\text{OH})_3$ and FePO_4 . After recovering Cu and Ni-Co alloys via electrodeposition, the pH of the solution was adjusted again to separate Mn and Li. This work provides new insights for the leaching research of ternary DESs.

Owing to its advantages, including high leaching efficiency, high selectivity, high ion transport efficiency, low melting point, biodegradability, low volatility, and low cost, DES deserves further exploration by researchers to investigate its leaching mechanism. However, it should be noted that the high viscosity of DESs can affect the leaching rate, and the stability of DESs during multiple cycles of reuse also needs to be considered.

In Table 1, the principal parameters that govern leaching performance, such as lixiviant concentration, residence time, temperature, and solid-to-liquid (S/L) ratio, are collated. The optimal window is defined by a “three-low and one-high” metric: minimal reagent concentration, shortened duration, reduced temperature, and maximized S/L ratio. Moreover, leaching schemes that afford preferential dissolution of lithium are strongly advocated, as they curtail downstream separation stages and thereby compress the overall flowsheet.

This section details the application of hydrometallurgy in recycling S-LFP. Hydrometallurgy recovers valuable metals and resynthesizes new cathode materials by breaking down the structure of S-LFP. Although its leaching process is relatively short and the industrial technique is mature, it consumes large quantities of chemical reagents and poses a risk of equipment corrosion. Hydrometallurgy mainly includes acid leaching, non-acid leaching, and DES leaching. Among these, acid leaching offers high efficiency and low cost. However, the acidic wastewater and waste residues generated require additional treatment to avoid environmental pollution. Compared with acid leaching, acid-free leaching reduces the usage of acidic reagents and mitigates environmental pollution, aligning with the concept of green recycling. Nevertheless, the lack of acid corrosion results in a generally lower leaching rate of Li^+ . DES leaching is an emerging recycling method in recent years, featuring advantages such as green sustainability, high efficiency, and mild leaching conditions. However, since it is still in the research and development stage, its industrial application potential needs further improvement.

3.2 | Direct Regeneration

Direct regeneration differs from pyrometallurgy and hydrometallurgy in that it does not require decomposing S-LFP cathode materials to extract valuable metals for resynthesizing new cathode materials. Instead, direct regeneration focuses on repair, restoring the structure and addressing defects of material

TABLE 1 | Information on hydrometallurgical recovery of S-LFP.

Method	Reagent	Condition	Efficiency	Reference
Inorganic acid leaching	0.315 M $\text{H}_2\text{SO}_4 + \text{H}_2\text{O}_2$	S/L = 96.87 g L^{-1} 62°C 90 min	95.23% Li	[77]
	0.88 M H_3PO_4 LFP: NMC = 2:1	L/S = 33 L g^{-1} , 80°C 2 h	99.6% Li	[78]
Organic acid leaching	0.8 M $\text{HCl} + \text{IL-ABS}$	S/L = 40 g L^{-1} , 25°C 2 h	90% Li, 98% Fe	[82]
	0.8 M $\text{CH}_3\text{COOH} +$ 6 vol% H_2O_2	S/L = 120 g L^{-1} , 50°C 30 min	94.08% Li	[84]
	0.2 M LA + 5% H_2O_2	S/L = 40 g L^{-1} , 40°C 30 min	99% Li, 2% Fe	[85]
Acid-free leaching	Lemon juice + 6 vol% H_2O_2	S/L = 67 g L^{-1} , 25°C 90 min	94.83% Li, 4.05% Fe	[79]
	$\text{Na}_3\text{Cit} + 1 \text{ ml } \text{H}_2\text{O}_2$	500 rpm 5 h	98.9% Li, 5.1% Fe	[89]
	LFP: LCO = 1:1 0.15 M H_2SO_4	650 rpm 5 h, 90°C 1 h	99.9% Li, 88.6% Co	[90]
	$\text{NH}_4\text{Fe}(\text{SO}_4)_4$	S/L = 50 g L^{-1} , 50°C 30 min	97.8% Li	[91]
	PMS	S/L = 50 g L^{-1} , 60°C 50 min	96.79% Li	[92]
	7 M $\text{NaOH} + \text{Ca}(\text{OH})_2$	L/S = 100 g L^{-1} , 250°C 2 h	97.88% Li, 0.02% Fe	[93]
	15 vol% $\text{H}_2\text{O}_2 + \text{LiOH} + \text{CA}$	S/L = 10 g L^{-1} , 50°C 30 min 350°C 1 h, 650°C 10 h	97.6% Li	[95]
DES leaching	Air- H_2SO_4	550°C 1 h, 60°C 2 h	97.48% Li	[80]
	$\text{C}_2\text{H}_3\text{ClO}_2$: ethanol = 1:1	S/L = 10 g L^{-1} , 70°C 6 h	100% Li, 0.03% Fe	[81]
	ChCl: EG: maleic acid = 1:1.5:0.5	L/S = 50 mL g^{-1} , 80°C 2 h	99% Li, 99% Fe	[99]

through methods such as direct lithium supplementation [18]. In this section, we introduce four direct regeneration methods: solid-phase sintering method, hydrothermal method, electrochemical method, and DES method, then analyze their advantages and disadvantages, respectively.

3.2.1 | Solid-Phase Sintering Method

Solid-phase sintering refers to the process of mixing S-LFP cathode powder with an appropriate amount of lithium source, carbon source, and reducing agent, followed by high-temperature calcination under a protective atmosphere to restore the material's electrochemical performance [100, 101]. As the temperature increases, lithium from the lithium source diffuses into vacancies under the driving force of high temperature, effectively compensating for the lithium loss in S-LFP. Meanwhile, the added reducing agent reduces Fe^{3+} to Fe^{2+} , repairing the impurity phases in S-LFP [102, 103]. After the reaction, the carbon source can form a doped carbon layer or coating, which enhances the electronic conductivity and structural stability of materials [104].

Inorganic lithium salts are used as lithium sources in solid-phase sintering and are often combined with eutectic molten salt-assisted calcination for regeneration. Xiao et al. [105] selected LiOH and LiI as a binary molten salt system to achieve one-step closed-loop reduction of S-LFP. The mechanism diagram of direct regeneration in this work is shown in Figure 5A. During the heat treatment process, LiOH and LiI can form a uniform

molten environment at 200°C, enabling full contact between Li^+ and LFP particles. I^- reduces Fe^{3+} to Fe^{2+} at 450°C while lowering the activation energy barrier for Fe to migrate back to Fe sites, which helps repair Li-Fe anti-site defects [111]. The conversion of Fe^{3+} to Fe^{2+} also promotes the diffusion rate of Li^+ , and increasing temperature further facilitates lithium supplementation and structural repair. The sublimated iodine enters the collection device along with Ar, achieving a 100% recovery efficiency. The S-LFP repaired by this method exhibits a capacity retention rate of 95.7% after 300 cycles at 1C. To balance relithiation and the optimization of the structural morphology of the carbon coating, Sun et al. [112] proposed an “ice-fire” two-step template-assisted method for regenerating S-LFP. The “ice step” refers to a solution relithiation process using LiOH as the lithium source, glucose as the reducing agent, and NaCl as the templating agent, followed by freeze-drying overnight in a vacuum. Subsequently, the freeze-dried powder is subjected to high-temperature calcination. After washing off NaCl and excess Li, the repaired LFP is obtained. The “ice step” accurately supplements the lithium missing in S-LFP while reducing Fe^{3+} , and the solution environment provides a more uniform porous carbon coating for subsequent annealing. In the “fire step,” NaCl acts as a template to construct a 3D interconnected porous carbon network, which provides more Li^+ migration channels and also enables the control of carbon hybridization. With the increase in glucose content, the capacity of the R-LFP varies in the range of 150–170 mAh g^{-1} , showing a significant performance improvement. In addition, in order to repair severely degraded LFP

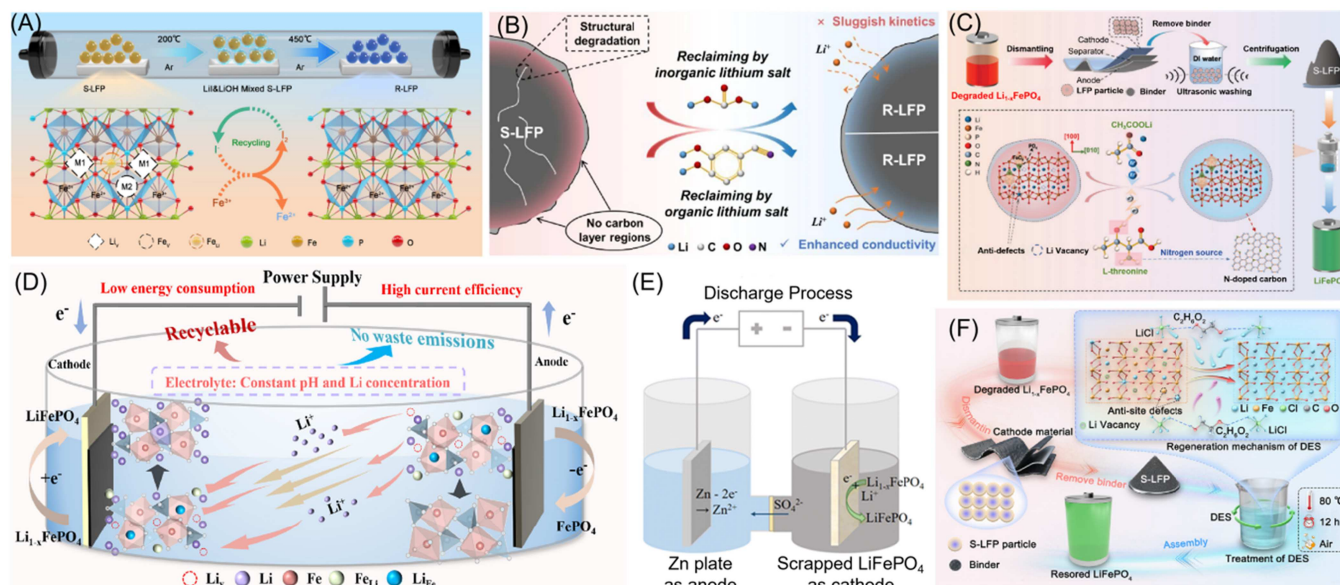


FIGURE 5 | The flowchart and mechanism diagram of direct regeneration: (A) Regeneration of S-LFP via a low-temperature molten salt system. Reproduced with permission: Copyright 2025, Elsevier [105]. (B) The comparison between using Li₂DHBN and inorganic salt. Reproduced with permission: Copyright 2023, Springer Nature [106]. (C) Regeneration of S-LFP using a hydrothermal reaction system composed of CH₃COOLi and L-threonine. Reproduced with permission: Copyright 2024, Wiley [107]. (D) Electrochemical regeneration method using S-LFP as cathode and anode. Reproduced with permission: Copyright 2024, Elsevier [108]. (E) Electrochemical regeneration method for relithiation of S-LFP in aqueous solution system. Reproduced with permission: Copyright 2022, Royal Society of Chemistry [109]. (F) Direct regeneration of S-LFP using DES composed of LiCl and EG. Reproduced with permission: Copyright 2025, Wiley [110].

(capacity $\leq 20\%$), Cheng et al. [113] proposed a novel method that constructs physical confinement in solid-phase sintering. Li₂CO₃ was added as the lithium source, and polyvinylpyrrolidone (PVP) was added as the reducing agent. The mixture was ball-milled with ethanol and then dried; the powder was pressed into blocks at a pressure of 30 MPa, then followed a sintering process. This process is called physically confined-domain pressure sintering. During the process, fragmented crystals recombine into complete ones, and the constructed Li⁺ transport channels promote Li⁺ diffusion. Meanwhile, PVP reduces Fe³⁺ to Fe²⁺ at high temperatures, eliminating the FP phase. The LFP with physical confinement introduced is effectively restored and exhibits improved fast-charging performance. Even at a high rate of 5C, it maintains an ultra-high capacity retention rate of 88.88% after 1500 cycles. This method is easy for large-scale batch production and provides a new solution for the direct repair and regeneration of industrial-grade S-LFP.

Inorganic lithium salts usually require additional reducing agents or carbon sources as auxiliaries, while suitable organic lithium salts can simultaneously act as lithium sources, reducing agents, and carbon sources, which makes them efficient multi-functional lithium salts. Ji et al. [106] synthesized a multi-functional organic lithium salt, 3,4-dihydroxybenzonitrile (Li₂DHBN). After mixing and grinding with S-LFP, the mixture was calcined under an Ar/H₂ atmosphere. The resulting R-LFP exhibited a capacity retention rate of 88% after 400 cycles at 5C. The functional groups of the organic lithium salt can effectively couple with S-LFP particles, facilitating the supplementation of active lithium. The reductive gas generated by the cyano group (—CN) at high temperatures reduces Fe³⁺ to Fe²⁺. Meanwhile, the carbon layer produced by pyrolysis coats the particle surface, improving the kinetic activity of Li⁺. The schematic diagram is shown in Figure 5B. This method can

repair LIBs with different degrees of degradation and different types, featuring an extremely wide application range. Moreover, organic lithium salts can form a carbon coating on S-LFP surfaces at low temperature, while high temperature promotes Li⁺ diffusion, realizing the effect of “low-temperature carbon coating and high-temperature lithium supplementation.” The reductive gas generated by specific functional groups can reduce the valence state of iron ions, providing a new direction for the regeneration of S-LFP via the solid-phase sintering method.

However, the regeneration process of solid-phase sintering inevitably requires high temperatures and high energy input, which consumes significant energy and results in substantial carbon emissions. This poses considerable risks to the environment and is inconsistent with the concept of low-carbon and green development. Additionally, solid-phase sintering involves a multiphase reaction, which is affected by factors such as the degree of mixing and the particle size of lithium salts. These factors lead to a low reaction interface area, low Li⁺ transport efficiency, and uneven interface reactions, consequently causing uneven Li⁺ supplementation and impairing the performance of regenerated batteries. These are currently the two primary challenges and difficulties in solid-phase sintering.

3.2.2 | Hydrothermal Method

The hydrothermal method typically uses deionized water as the medium for Li⁺ transport. A properly proportioned lithium source and reducing agent are added and mixed with S-LFP powder. By means of high-pressure conditions, the transport efficiency of Li⁺ is improved, thereby achieving the goal of repairing S-LFP. Sometimes, an additional annealing process is also required to repair the crystal structure [114]. Compared

with the solid-phase sintering method, the hydrothermal method has milder reaction conditions: the reaction at the solid–liquid two-phase interface is more uniform, and the reduction environment is more stable. High pressure can significantly reduce the ion diffusion energy barrier, effectively promoting Li^+ diffusion [115, 116]. In addition, during the hydrothermal regeneration process, carbon emissions are lower than those of the solid-phase method, and the crystal morphology is controllable, while there is no need to consider the Li/Fe ratio [117].

To address the problem of the single function of reducing agents, Tang et al. [107] used L-threonine, a natural multifunctional substance, as the reducing agent. Its hydroxyl groups ($-\text{OH}$) act as electron donors, which can reduce lithium vacancies and eliminate anti-site defects, its amino groups ($-\text{NH}_2$) are used for modifying the carbon conductive layer, and nitrogen doping through these groups creates more active sites and improves conductivity [118]. The mechanism and flow chart of this work are shown in Figure 5C. After hydrothermal treatment, the R-LFP exhibited a high discharge capacity of 147.9 mAh g^{-1} at 1 C and maintained a capacity retention rate of 86% after 500 cycles at a high rate of 5 C. This method is also applicable to the regeneration of severely degraded materials. Only an additional annealing step is required to repair the fragmented crystal structure. The repaired black mass also exhibits excellent electrochemical performance. To achieve large-scale repair of S-LFP using a single effective reagent, Feng et al. [119] proposed an in situ particle reconstruction strategy for the direct regeneration of industrial-grade LFP black mass. Glycerol was selected as the reducing agent and solvent, and LiOH was selected as the lithium source. Subject these mixtures to hydrothermal and annealing treatment; the hydroxyl groups ($-\text{OH}$) abundant in glycerol provide electrons to form a reductive environment, which can reduce Fe^{3+} and Li-Fe anti-site defects. The high viscosity of glycerol enables it to significantly inhibit the Oswald ripening effect when used as a solvent. This allows fragmented small particles to reaggregate into complete particles. The regenerated black mass obtained in this work exhibited a discharge capacity of 123.2 mAh g^{-1} at 5 C, with a capacity retention rate of 93.1% after 500 cycles. In addition to common organic reducing agents, Jiang et al. [120] compared the reducibility of several common organic reducing agents (citric acid, tartaric acid, and ascorbic acid) and inorganic reducing agents (H_2SO_3 , Na_2SO_3). They found that ascorbic acid exhibited the strongest reducibility and could complete the relithiation of S-LFP under mild hydrothermal conditions at 80°C . However, organic reducing agents typically generate complex by-products. When the neutral reducing agent Na_2SO_3 is combined with lithium salts, the solution containing excess Na^+ requires further treatment [121]. In contrast, the only by-product generated by the inorganic acidic reducing agent H_2SO_3 is SO_4^{2-} . After using H_2SO_3 as the reducing agent, the R-LFP achieved an initial discharge capacity of 142.5 mAh g^{-1} at 0.1 C, with a capacity retention rate of 98.9% after 100 cycles. Excess lithium can be purified by adjusting the pH value, and Na_2CO_3 can be added for precipitation and recovery.

The hydrothermal method can provide a mild and uniform lithium-supplementation environment for S-LFP while avoiding the adverse effects of long-term heat treatment in solid-phase calcination on particle morphology and size

[122]. However, the hydrothermal method has strict requirements for reaction control. It requires high-pressure reaction conditions, which impose high demands on equipment. In addition, since the reaction occurs in a liquid phase, a large amount of wastewater is inevitably generated, so this places high requirements on safety and environmental protection.

3.2.3 | Electrochemical Method

As an emerging direct regeneration method, the electrochemical method adopts a three-electrode system or a H-type electrolytic cell. It uses S-LFP as the working cathode and metallic Li as the anode and applies a voltage to drive Li^+ in the electrolyte to migrate directly into the lithium vacancies of the cathode, thereby achieving the goal of relithiation [123, 124].

Due to the characteristic of S-LFP that it has a built-in lithium source, Yang et al. [108] used S-LFP materials as both the cathode and anode in an electrolytic system, enabling lithium ions to deintercalate from the anode during charge–discharge to intercalate into the cathode, thereby directly regenerating LFP. Based on the ICP-OES results of the cathode material, the mass of the anode material was calculated by combining the Li-Fe molar ratio, achieving precise lithium addition. At the optimal voltage (1 V), the anode was delithiated to form FePO_4 , and the extracted Li^+ then intercalated into the lithium vacancies of the cathode through the electrolyte to form LiFePO_4 , completing the regeneration (Figure 5D). The R-LFP exhibited an excellent high initial capacity (136.5 mAh g^{-1} at 1 C) and outstanding cycling stability (a capacity retention rate of 95.32% after 300 cycles at 5 C). Considering the green economy of aqueous relithiation, Zhou et al. [109] selected a H-type electrolytic cell for S-LFP regeneration with an anion exchange membrane separating the cathode and anode. The cathode compartment contained an S-LFP suspension (S-LFP powder uniformly mixed into a Li_2SO_4 solution), and the anode was a zinc plate (immersed in a ZnSO_4 solution), as shown in Figure 5E. At a current of 5 mA, the zinc plate undergoes an oxidation reaction ($\text{Zn} - 2\text{e}^- = \text{Zn}^{2+}$), while the S-LFP in the cathode gains electrons and undergoes a reduction reaction ($\text{Li}_{1-x}\text{FePO}_4 + x\text{Li}^+ + x\text{e}^- = \text{LiFePO}_4$) to form LFP. The LFP obtained via this reaction not only exhibits good crystallinity and high lithium-supplementation efficiency but also achieves a high initial capacity of 134 mAh g^{-1} at 1 C, with a capacity retention rate of 85.5% after 300 cycles. Currently, few studies can directly repair S-LFP electrode sheets without damaging them. Against this background, Fan et al. [125] proposed a novel in situ regeneration strategy based on a functionalized prelithiated separator (FPS). In this study, the FPS was fabricated simply by coating a thin layer of $\text{Li}_2\text{C}_2\text{O}_4$ /mesoporous carbon (CMK-3) composite onto a commercial separator. S-LFP electrode slices were directly assembled into a full cell with the FPS and a fresh graphite anode, eliminating the need for pretreatment of the spent cathode material. $\text{Li}_2\text{C}_2\text{O}_4$ was used as the lithium source in this work due to its characteristic high irreversible capacity ($> 500 \text{ mAh g}^{-1}$). However, its relatively high decomposition potential ($\sim 4.7 \text{ V}$) severely limits its application as a lithium-supplementing agent. Therefore, it was composited with CMK-3, which has a high specific surface area ($800 \text{ m}^2 \text{ g}^{-1}$) and high electronic conductivity, to reduce the decomposition potential

to approximately 4.4 V. During the first charging process, $\text{Li}_2\text{C}_2\text{O}_4$ undergoes irreversible electrochemical decomposition to generate Li^+ and CO_2 . These Li^+ intercalate into the lithium-deficient cathode during the discharge process, thereby repairing the S-LFP. CO_2 is removed through a degassing process before cycling, and the pores generated by the decomposition of $\text{Li}_2\text{C}_2\text{O}_4$ also facilitate electrolyte penetration. The assembled cell exhibited a specific capacity of 146.7 mAh g^{-1} and maintained a high capacity retention rate of 90.7% after 292 cycles. Moreover, after long-term cycling at high rates, the capacity recovered rapidly once the rate was reduced to 0.1 C.

Although the electrochemical method does not require high-pressure or high-temperature conditions, it demands strict control over current/voltage parameters, because excessively high values can lead to fragile and brittle crystal structures. More importantly, it has high requirements for raw materials, involves a long preparation process, and faces significant challenges in large-scale promotion. Therefore, the electrochemical method might not be suitable for large-scale industrial applications, and additional research is needed to advance its practical implementation.

3.2.4 | Deep Eutectic Solvent Regeneration

DESs are a new type of green solvent that has attracted considerable attention in recent years. Composed of a HBA and a HBD, the hydrogen bond interaction between HBA and HBD can effectively lower the eutectic point of DESs, enabling them to directly repair spent LIBs at a low temperature [126]. Initially, DESs were used to dissolve spent cathode materials and extract valuable metals [127–129]. When applied in direct regeneration processes, lithium salts are often selected as HBAs to provide active lithium, while reagents with excellent Li^+ transport performance (such as H_2O , ethanol, ethylene glycol, and glycerol) are used as HBDs [126]. Additionally, the reductive environment created by HBDs can promote the reduction of Fe^{3+} to Fe^{2+} and correct Li-Fe anti-site defects.

Since lithium-containing DES can significantly repair S-LFP, Lin et al. [110] selected LiCl as HBA and ethylene glycol as HBD. After these two components form a DES solution, an appropriate amount of S-LFP is added. The reaction was conducted at a low temperature (80°C) for 12 h. During the reaction process (Figure 5F), the DES acts as a direct lithium source to supplement the lithium missing in S-LFP. The hydroxyl groups ($-\text{OH}$) of ethylene glycol serve as electron donors to reduce Fe^{3+} . Meanwhile, they lower the energy barrier for iron ions to return to their original lattice sites, reducing Li-Fe anti-site defects. The resulting LFP delivered an initial discharge capacity of 155.6 mAh g^{-1} at 0.1 C, and its capacity remained at 93% of the initial value after 300 cycles at 1 C. This method avoids high-temperature calcination, can be scaled up for production, and has high economic and environmental value. Moreover, ternary DES is also commonly used in the regeneration of S-LFP. Pu et al. [130] used a DES composed of LiI , urea, and ascorbic acid (designated as IUA) as the regeneration solvent for S-LFP. During the reaction, LiI creates a lithium-rich environment, ascorbic acid provides a reductive environment, and urea acts as a stable medium for Li^+ transport. These components simultaneously supplement the lithium lost in S-LFP, reduce the valence state of Fe^{3+} , and minimize Li-Fe

anti-site defects. The R-LFP exhibited excellent electrochemical performance: it delivered a high discharge capacity of 160 mAh g^{-1} at 0.1 C and maintained 85.8% of its capacity after 400 cycles at 1 C. Furthermore, the IUA solvent could be collected and reused for a second round of regeneration. In this case, the R-LFP still achieved a discharge capacity of 159 mAh g^{-1} at 0.1 C, showing little difference from the LFP regenerated in the first round. To investigate the feasibility of repairing S-LFP by DES at room temperature, Yingnakorn et al. [131] used a DES composed of lithium acetate and ethylene glycol, with an appropriate organic reducing agent added, to directly relithiate S-LFP. They compared the reduction effects of five organic reducing agents on S-LFP. It was found that only hydroquinone exhibited reversibility in addition to strong redox properties. This indicates that hydroquinone is a recyclable reducing agent, which can be reused and thus holds high economic and environmental protection value. Inductively coupled plasma mass spectrometry detection showed that the atomic ratio of the regenerated material was $\text{Li:Fe:P} = 1:1:1.02$, confirming the success of relithiation. The R-LFP delivered a discharge capacity of 153.9 mAh g^{-1} at 0.1 C. Moreover, after long-term cycling, when the rate was restored to 0.1 C, its capacity recovered to 151.4 mAh g^{-1} , demonstrating excellent reversibility.

It can be observed that DES exhibits advantages such as low melting points, high solvating power, and environmental benignity. The chemical reaction kinetics and molecular dynamics of DES can be regulated by controlling the proportion of HBA and HBD. However, it is important to note that the high viscosity of DES may impede molecular diffusion, and the cost of certain DES components is relatively high. Addressing these issues remains a crucial research direction at present.

In Table 2, the principal parameters that govern regeneration performance, such as temperature, reagent selection, reaction duration, voltage, and current intensity, are collated. Among these, low-cost reagents, low-temperature, and short-duration reaction conditions are the optimal process choices. For the electrochemical method, the current or voltage density directly affects the de-intercalation rate of Li^+ , and the electrolysis duration directly influences the Li^+ replenishment effect and material structural stability. Therefore, it is necessary to select appropriate electrochemical parameters.

This section details the application of the direct regeneration method in recycling S-LFP. Direct regeneration can repair S-LFP without damaging the structure of the cathode material, restoring its performance to the initial state. Among direct regeneration techniques, the solid-phase sintering method features a simple process and easy operation. However, its high-temperature and high-energy consumption characteristics are harmful to the environment, and it is difficult to precisely control the crystal structure and composition. The hydrothermal method enables lithium replenishment, Fe^{3+} reduction, and precise regulation of material morphology and particle size in a mild liquid-phase environment. Nevertheless, the high-pressure conditions are unfavorable for large-scale production, and the liquid-phase environment also generates a large amount of wastewater that requires additional treatment. In contrast, the electrochemical method can achieve Li replenishment without high-temperature or high-pressure conditions. However, this method is only suitable for cathode materials

TABLE 2 | Information on direct regeneration method of S-LFP.

Method	Reagent	Condition	Performance	Reference
Solid-phase sintering method	S-LFP + LiOH + LiI	200°C 4 h, 450°C 4 h	95.7% (300r, 1 C)	[105]
	S-LFP + LiOH + Glucose + NaCl + Urea	60°C 2 h, 650°C 3 h	141.79 mAh g ⁻¹ , 95.7% (200r, 1 C)	[112]
	S-LFP + Li ₂ CO ₃ + 10 wt% PVP	600 rpm 6 h, 750°C 10 h	88.8% (1500r, 5 C)	[113]
	S-LFP + 5 wt% Li ₂ DHBN	800°C 6 h	88% (400r, 5 C)	[106]
Hydrothermal method	S-LFP + CH ₃ COOLi + L-threonine + H ₂ O	180°C 6 h	147.9 mAh g ⁻¹ (1 C), 86% (500r, 5 C)	[107]
	S-BM + LiOH + Glycerol	180°C 10 h, 600°C 3 h	123.2 mAh g ⁻¹ , 93.1% (500r, 5 C)	[119]
	S-LFP + LiOH + H ₂ SO ₃ + H ₂ O	90°C 6 h	142.5 mAh g ⁻¹ , 98.9% (100r, 0.1 C)	[120]
	Cathode and anode: S-LFP Electrolyte: Li ₂ SO ₄	Voltage: 1 V	136.5 mAh g ⁻¹ (1 C), 95.32% (300r, 5 C)	[108]
Electrochemical method	Cathode: Zinc plate Electrolyte: ZnSO ₄	H-type electrolytic cell Current: 5 mA	134 mAh g ⁻¹ , 85.5% (300r, 1 C)	[109]
	Anode: S-LFP suspension Electrolyte: Li ₂ SO ₄	TIA: 150%		
	Cathode: Graphite Anode: S-LFP Electrolyte: LiPF ₆ Separator: Li ₂ C ₂ O ₄ /CMK-3	Ultrasonic blending and recrystallization synthesis Li ₂ C ₂ O ₄ /CMK-3	90.7% (292r, 1 C)	[125]
	S-LFP + LiCl + EG	DES: 120°C 20 min, 80°C 12 h	155.6 mAh g ⁻¹ (0.1 C), 93% (300r, 1 C)	[110]
DES	LiI + Urea + AA	DES: 80°C 12 h, 100°C 10 h	160 mAh g ⁻¹ (0.1 C), 85.8% (400r, 1 C)	[130]
	S-LFP + CH ₃ COOLi·H ₂ O + EG + Hydroquinone	DES: 60°C, 25°C 1–3 h	153.9 mAh g ⁻¹ (0.1 C)	[131]

with minor damage and requires precise control of conditions such as current and voltage, making it unsuitable for industrial production. DES can regenerate S-LFP under atmospheric pressure and low-temperature conditions, and the regenerated DES solvents can be recovered and recycled to improve resource utilization. However, factors such as the high viscosity of DES and the high cost of some components hinder its industrial application.

4 | Upcycling Method of S-LFP

The upcycling of spent cathode materials aims to upcycle and reprocess S-LFP into high-performance materials, thereby enhancing the economic added value of regenerated products. It is not limited to the reuse of resources and performance recovery but rather involves the upcycling of spent batteries to obtain electrode materials with superior performance and function or other materials with higher economic value [132, 133]. This section introduces the upcycling methods of LFP in three parts: structural optimization to regenerate high-performance LFP through strategies such as doping and coating, upcycling of LFP into LFMP via phase transformation, and upgrading and reprocessing into advanced post-metal-ion batteries or catalytic materials.

4.1 | Optimizing the Structure to Generate High-Performance LFP

Structural optimization is a common method for upcycling S-LFP batteries. This strategy aims to improve the interfacial performance and structural stability of LFP by introducing specific elements and applying carbon coating, thereby enhancing the electrochemical performance of the batteries [134–137]. The carbon coating layer on the surface of LFP particles can improve electronic conductivity, while element doping can achieve the dual effects of reducing intrinsic defects and optimizing the lattice parameters of the electronic structure [138]. This method is usually combined with other direct regeneration techniques, forming a uniform coating layer during the annealing process. The element-doped carbon coating can effectively reduce the diffusion barrier of lithium ions at the interface and enhance electronic conductivity as well as the structural stability of LFP.

A primary strategy for structural optimization in the upcycling of S-LFP batteries involves either doping the bulk structure of LFP or optimizing the coating layer. Xiao et al. [139] used polyacrylonitrile (PAN) as a reducing agent and additive and lithium acetate as the lithium source to repair LFP (Figure 6A). The mixture of S-LFP, PAN, and CH₃COOLi was blended with

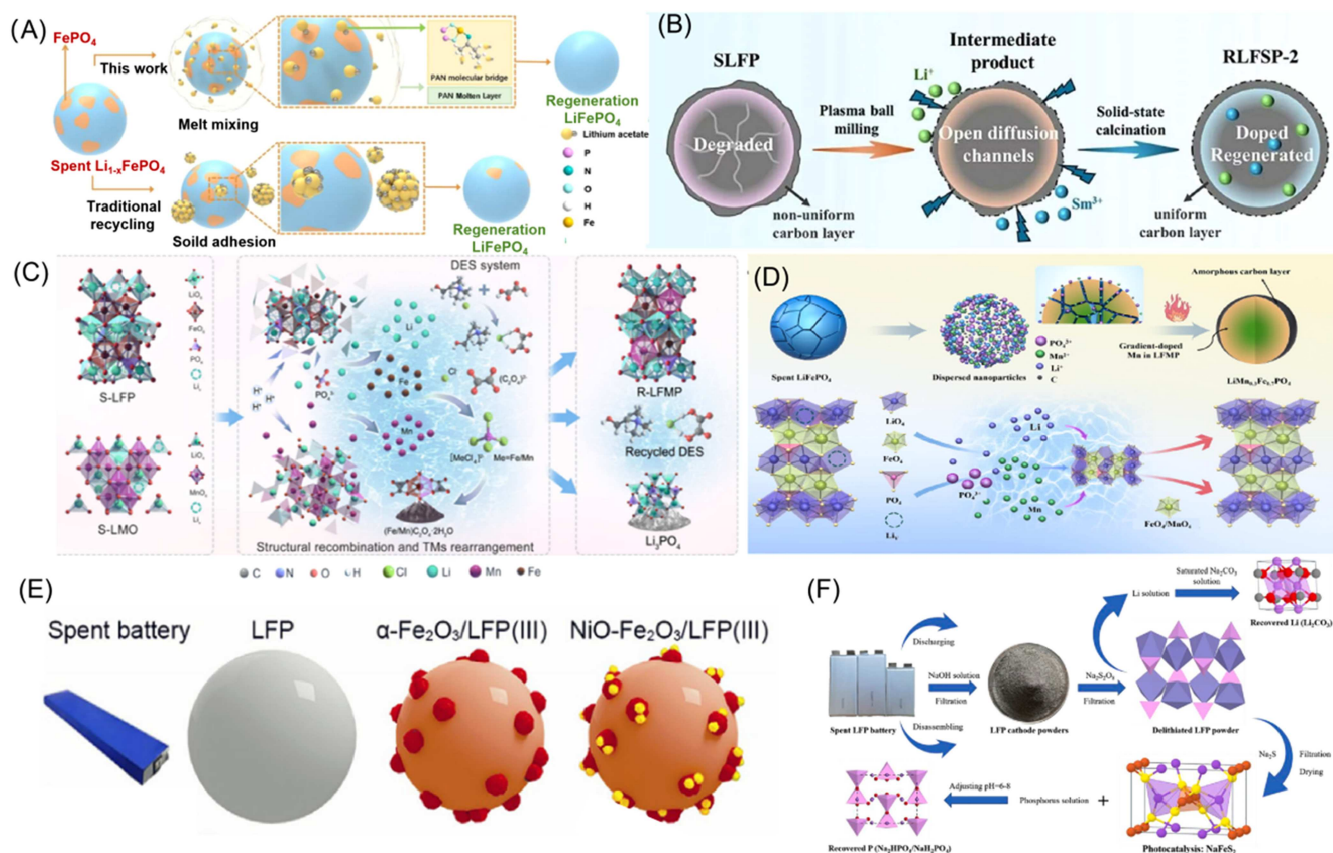


FIGURE 6 | The flowchart and mechanism diagram of upcycling: (A) Comparison chart of this work using PAN as nitrogen source versus traditional recycling method. Reproduced with permission: Copyright 2025, American Chemical Society [139]. (B) Regeneration of S-LFP by Sm doping via plasma ball milling-assisted calcination. Reproduced with permission: Copyright 2025, Tsinghua University Press [140]. (C) Structural change of S-LFP achieved by dissolving and reconstructing with DES composed of ChCl and OA. Reproduced with permission: Copyright 2024, Springer Nature [141]. (D) Upcycling of S-LFP into LFMP via defect inheritance strategy. Reproduced with permission: Copyright 2025, Elsevier BV [142]. (E) Conversion of S-LFP into a high-performance OER electrocatalyst via direct oxidation. Reproduced with permission: Copyright 2025, John Wiley and Sons Ltd [143]. (F) Using $\text{Na}_2\text{S}_2\text{O}_8$ to extract Li and P in S-LFP for the resynthesis of the photocatalyst NaFeS_2 . Reproduced with permission: Copyright 2022, Elsevier [144].

glucose and ethanol. After being uniformly mixed by high-speed ball milling, the mixture undergoes a three-stage annealing treatment. The uniform medium layer formed by PAN at low temperatures enhances the contact between CH_3COOLi and S-LFP, facilitating the supplementation of Li^+ . At high temperatures, PAN decomposes to generate a reductive atmosphere, which promotes the reduction of Fe^{3+} to Fe^{2+} . Meanwhile, the uniform N-doped carbon layer formed by pyrolysis improves the conductivity of the cathode material. Under these conditions, the R-LFP exhibits a high capacity of 155 mAh g^{-1} at 0.1 C, and after 400 cycles at 1 C, the capacity retention rate is approximately 87%. Since plasma ball milling can effectively activate the material surface, Li et al. [140] proposed to use it to assist in the solid-phase sintering regeneration of S-LFP. An appropriate amount of Li_2CO_3 was added to compensate for the lithium deficiency of S-LFP, as determined by ICP-OES. Then, samarium(III) nitrate hexahydrate ($\text{Sm}(\text{NO}_3)_6 \cdot 6\text{H}_2\text{O}$) was incorporated for Sm doping. After the high-speed plasma ball mill, the mixture was placed to anneal. The mechanism diagram of this work is shown in Figure 6B. During the plasma ball milling process, high-energy plasma induces mechanochemical activation, which effectively exposes the material surface and facilitates Sm doping. The relatively

large ionic radius of Sm can open up Li^+ transport channels and increase the diffusion rate [145]. Meanwhile, Sm doping expands the unit cell volume, which helps alleviate volume shrinkage during long-term cycling and stabilize the crystal structure. Calcination removes most impurities in the material, and the residual carbon in S-LFP undergoes an in situ carbothermal reduction reaction, promoting the reduction of Fe^{3+} to Fe^{2+} . At the same time, the carbon layer is redistributed to form a uniform Sm-containing conductive network. The LFP repaired in the above work exhibited a capacity of 142.2 mAh g^{-1} at 1 C and a capacity retention rate of 96.5% after 200 cycles. This confirms that the R-LFP has a more stable structure and a higher Li^+ diffusion rate. Considering during the pretreatment of S-LFP, Cu impurities will be inevitably introduced into the samples. It is important to study the effect of the amount of Cu on the regeneration of S-LFP. Through detailed density functional theory (DFT) calculations, Zeng et al. [146] found that appropriate Cu doping improves Li^+ diffusion ability and enhances electronic conductivity. CuO and S-LFP were mixed at a ratio of 200 ppm:1. The mixture was heated in air to remove the binder and conductive agent, during which LFP was oxidized to $\text{Li}_3\text{Fe}_2(\text{PO}_4)_3$ and Fe_2O_3 . Then Li_2CO_3 was added as the lithium source, and glucose was added

as the reducing agent. After thorough mixing, heat treatment was performed. In this process, Cu was effectively doped into the lattice, which weakened the Li-O bond interaction, thereby increasing the Li⁺ diffusion rate and suppressing Li-Fe anti-site defects [147]. The R-LFP achieved a capacity of 130 mAh g⁻¹ at 2 C, with a capacity retention rate of 100% after 300 cycles. The full cell assembled with this R-LFP exhibited a capacity of 128 mAh g⁻¹ at 1 C. To reconstruct the damaged carbon layer, Liu et al. [148] utilized natural tea polyphenols as electron donors and AlPO₄/Li₃PO₄ (AP/LP) while introducing Al doping, to upcycle LFP materials. The specific process is divided into two steps: repair and regeneration. First, S-LFP was dispersed into a hydrothermal repair solution consisting of LiOH and tea polyphenols. Then, the hydrothermally repaired powder was mixed with Al(NO₃)₃/NH₄H₂PO₄, excess Li₂CO₃ was added, and annealing was done to obtain the upcycled LFP powder. During the hydrothermal repair process, tea polyphenols provide electrons to reduce Fe³⁺ and synergize with Li⁺ from LiOH to compensate for lithium and eliminate Li-Fe anti-site defects. Al diffuses into the LFP lattice and replaces Fe sites during annealing, strengthening Fe-O bonds. The generated AP/LP hybrid patch coating repairs the carbon layer and performs targeted surface reconstruction. The assembled full cell exhibits an initial capacity of 124.3 mAh g⁻¹ at 2 C, with a capacity retention rate of 92.1% after 400 cycles, proving that this work significantly improves the cycling stability of LFP.

Structural optimization can significantly improve the electrochemical performance of LFP; however, additional attention must be paid to the type and dosage of doping elements. When screening doping elements, DFT can be used to optimize the doped structure and calculate key information such as lithium ion transport barriers, unit cell parameters, and energy band structures. This avoids aimless selection from the periodic table of elements. It is necessary to select appropriate elements for doping and coating modification to prevent the occurrence of side reactions. Excessive doping may damage the original crystal structure of LFP and may also lead to uneven or excessively thick coating layers, which affect electronic conductivity. These challenges require further research and exploration to enable the broader application of structural optimization.

4.2 | Changing the Phase to Generate LFMP

Current regeneration methods focus on capacity recovery rather than component upgrading [149]. Given that the energy density of LFP (180 Wh kg⁻¹) is much lower than that of ternary batteries (250 Wh kg⁻¹) and the operating voltage of LFP is only 3.2 V, improving the median voltage is key to enhancing energy density. Since the operating voltage of LFMP ranges from 3.5 to 4.1 V, replacing part of the Fe sites with the transition metal Mn to increase the operating potential of LFP is an effective approach to improve the energy density of LFP [150–152].

The upcycling of mixed spent cathode materials (LFP and LMO) into high-performance LFMP not only enables the simultaneous recycling of these two types of spent cathodes but also introduces Mn into LFP through phase transformation to enhance the energy density. This addresses two key challenges: impure recycled battery raw materials and low energy density of regenerated batteries. Ji et al. [141] utilized an environmentally

friendly DES to simultaneously dissolve and reorganize the mixed S-LFP and spent LMO (S-LMO) materials and combined this with annealing treatment to reconstruct the crystal structure. Choline chloride was mixed with oxalic acid, and the mixture was heated until it turned into a liquid. S-LFP and S-LMO were then added to this liquid for leaching, resulting in the formation of Fe/MnCl₄²⁻ complexes. H₂O was added to adjust the viscosity of DES, converting the complexes into (Fe,Mn)C₂O₄·2H₂O precipitates, which act as the precursor of LFMP. The precursor was mixed with NH₄H₂PO₄ and LiOH, followed by ball milling and two-stage annealing treatment. Its schematic diagram is shown in Figure 6C; DES serves as a regulatory carrier for the composition and structure of the cathode material. Hydrogen in oxalic acid disrupts the structures of LFP and LMO, dissolving the Li, Mn, and Fe metals contained therein. Meanwhile, choline chloride and C₂O₄²⁻ reduce Mn⁴⁺ and Fe³⁺ to Mn²⁺ and Fe²⁺. This method yields a solid solution with 89.6% Fe and 82.2% Mn; approximately 96.7% of Li and 97% of P are retained in the filtrate and finally recovered in the form of Li₃PO₄. During the annealing process, oxalate ions form a carbon coating with a thickness of 4–5 nm. Fe and Mn are uniformly distributed on the surface and inside the particles, and Mn replaces Fe sites to form LiFe_{0.5}Mn_{0.5}PO₄, which enhances structural stability and electrical conductivity. The median voltage of the regenerated LFMP is increased to 3.68 V, with an initial capacity of 152 mAh g⁻¹ at 0.1 C. Its energy density (559 Wh kg⁻¹) is significantly higher than that of commercial LFP. Furthermore, after 1000 cycles at 1 C, 2 C, and 5 C, the capacity retention rates are 87%, 73%, and 75%, respectively, indicating stable long-term cycling performance. Utilizing the inherent structural defects of S-LFP (Li vacancies, cracks, lattice distortions), Ji et al. [142] converted these defects into transport channels for Li⁺/Mn²⁺, thereby achieving the supplementation of active lithium and gradient doping of Mn. Firstly, a mixture of S-LFP, Li₂CO₃, MnCO₃, NH₄H₂PO₄, and glucose was placed for mechanochemical ball milling. This process refines the particle size and converts microcracks into a nanoparticle network, which facilitates the rapid diffusion of Li⁺/Mn²⁺. Subsequently, the ball-milled mixture underwent carbothermal reduction and high-temperature lithium supplementation under an inert atmosphere. The sintering process during carbothermal reduction promotes Li⁺ compensation and enables the formation of Fe/Mn atomic solid solution. Meanwhile, a carbon coating is generated to enhance electrical conductivity and inhibit harmful interfacial reactions that may occur during long-term cycling (the specific mechanism is shown in Figure 6D). Gradient Mn doping can suppress Jahn-Teller distortion, prevent structural collapse, and significantly improve the cycling stability of LFMP. The LFMP regenerated via this defect inheritance mechanism exhibited a capacity retention rate of 83% after 900 cycles at 0.5 C, with an energy density as high as 563.5 Wh kg⁻¹. Combining hydrometallurgy and direct regeneration method, Lei et al. [153] proposed to leach Li/Fe/P from S-LFP and Li/Mn from S-LMO, then exploited the hydrothermal method to regenerate LFMP, and finally performed calcination to form carbon layer. First, they used sulfuric acid to leach 100% of Li/Fe/Mn/P. Then, ascorbic acid, Li/Fe/Mn/P sources and ethanol, were added, followed by a hydrothermal process. During this process, the hydrogen bonds of ethanol preferentially adsorbed to the oxygen sites on the particle surface, leading to the shortening of Mn-O bonds

and a reduction in crystal volume (resulting in a more stable structure). At the same time, Li–O bonds were lengthened, which facilitated the de-intercalation of Li⁺. During the calcination, a carbon layer was formed to improve the electrical conductivity of LFMP. The LFMP exhibited excellent rate performance and cycling stability: its capacity is 144.2 mAh g^{−1} at 1 C, and capacity retention rate is 87% after 1000 cycles. Even at a high rate of 5 C, the capacity retention rate remained 86.4% after 1000 cycles. Based on the challenge of controlling the uniformity of Li and Mn, Cheng et al. [154] proposed a solvothermal upcycling strategy to form LFMP with uniform element distribution through atomic-level mixing of Fe and Mn. They first completely dissolved S-LFP using sulfuric acid, then added ascorbic acid, manganese carbonate, and ammonium dihydrogen phosphate to introduce Mn for achieving atomic-level mixing of Fe and Mn. The mixture was homogenized with a polyethylene glycol solution for hydrothermal reaction. The treated sample was then mixed with PVP and subjected to high-temperature calcination to obtain LFMP. The atomic-level mixing of Fe and Mn results in uniform distribution of Fe and Mn in LFMP, enhancing structural stability. Mn doping also induces lattice expansion in LFMP, providing smoother diffusion channels for Li⁺. Meanwhile, PVP forms a carbon layer during the calcination process, which enhances the electronic conductivity of the material and promotes Li⁺ diffusion. The LFMP obtained through this method exhibits a capacity retention rate of 83.8% after 2500 cycles at 5 C, with an energy density of 561 Wh kg^{−1}, showing significantly improved electrochemical performance.

Upcycling regenerates S-LFP into high-energy-density LFMP via phase transformation is one of the current research hotspots. However, it should be noted that controlling the uniform distribution of Fe/Mn in LFMP and mitigating the Jahn-Teller effect caused by Mn introduction are key challenges in current research. Excessive Mn may reduce the specific discharge capacity of the cathode material and may also lead to lattice distortion or changes in electronic conductivity.

4.3 | Post-Metal-Ion Batteries and Catalytic Materials From Upcycling S-LFP

The primary manifestation of LFP failure is the loss of active lithium. However, recent research progress on sodium-ion batteries (SIBs) indicates that vacancy engineering can improve electrochemical performance to some extent. It alleviates structural collapse of sodium-storage electrode materials during rapid charge-discharge processes, and further promotes charge transfer while reducing volume strain [155–157]. Additionally, studies have shown that iron-based derivatives (such as FePO₄ and Fe₂O₃) generated after LFP failure exhibit catalytic activity similar to the Fenton reaction. The Fenton-like reaction can efficiently degrade refractory organic pollutants in industrial wastewater and maintain high catalytic efficiency under neutral or weakly acidic conditions. Thus, they can be converted into catalytic materials to extend the service life of LFP [158]. Moreover, the ferrite magnetic functional materials produced from these derivatives, which exhibit excellent properties such as high magnetic permeability, low loss, and wide frequency bandwidth, can be widely applied in many fields such as radar and microwave applications [159]. This type of upcycling is no

longer limited to traditional battery manufacturing, but extends to high-value-added products. These transformative upcycling routes are also current research hotspots.

S-LFP can be converted into an OER catalyst. Wang et al. [160] proposed a direct oxidative conversion strategy, which transforms S-LFP cathode material into high-performance OER electrocatalyst through a two-step air oxidation process (Figure 6E). In the first step, the S-LFP electrode material was calcined. This process not only stripped off the aluminum foil but also oxidized LFP into Li₃Fe₂(PO₄)₃ (LFP(III)). During this stage, uniformly distributed α-Fe₂O₃ nanodots were spontaneously formed on the particle surface, ensuring structural integrity while providing moderate OER activity. In the second step, LFP(III) was mixed with nickel acetylacetonate (Ni(acac)₂) and dispersed in ethanol. The mixture was subjected to ultrasonic treatment and then dried. At this point, NiO grew selectively on the α-Fe₂O₃ nanodots, generating an LFP(III)/NiO composite. The formed NiO/α-Fe₂O₃ nano-heterojunction could improve the interfacial charge distribution and accelerate charge transfer. The resulting catalyst only required overpotentials of 268 and 292 mV to achieve OER currents of 10 and 100 mA cm^{−2}, respectively. Furthermore, the phosphate rich LFP(III) could effectively inhibit Cl[−] adsorption, solving the problem that traditional catalysts are prone to corrosion in saline environments. Based on the defect inheritance strategy, Wang et al. [143] proposed utilizing the inherent structural defects of S-LFP for the design of SIB cathode materials. They separated Li and Fe to generate FePO₄ precipitates, which were then used for remanufacturing SIB cathodes, specifically to prepare the SIB cathode material Na₂FeP₂O₇ (NFPO-R). During the selective extraction of Li, Fe vacancies were retained in FePO₄. Thus, NFPO-R exhibits lattice distortion and volume expansion, which help widen the Na⁺ transport channels. The presence of vacancies also shortens Fe–O bonds, strengthens bonding interactions, and enhances the structural stability of NFPO-R. Additionally, electron accumulation occurs near Fe vacancies, improving the electronic conductivity of NFPO-R. The as-prepared NFPO cathode can be charged to 80% state in 6 min and maintains a capacity retention rate of 82.9% after 4000 cycles at a high rate of 10 C. Converting S-LFP into a photocatalyst is also one of the approaches to enhance the recycling value of spent batteries. Yue et al. [144] developed a facile method to remanufacture photocatalysts with synergistic adsorption-photocatalysis capabilities from S-LFP, while simultaneously recovering Li and P. The schematic diagram of this work is shown in Figure 6F. Na₂S₂O₈ is used to leach Li from S-LFP at room temperature. Under optimal conditions, nearly 100% of Li can be recovered. After drying, the delithiated LFP and Na₂S are mixed at a mass ratio of 1: 1 at room temperature to prepare NaFeS₂, enabling simultaneous recovery of 99.98% of P. NaFeS₂ exhibits a rough surface and loose structure, leading to a significant enhancement in adsorption capacity and photocatalytic activity. In addition, the heterostructure formed by multivalent Fe and S in NaFeS₂ endows it with a broad light absorption range (200–800 nm) and strong visible-light absorption capability [161]. Consequently, NaFeS₂ achieves a higher photocurrent density under visible light, which also accounts for its strong charge transfer efficiency. Moreover, its low photoluminescence intensity can significantly improve its photodegradation efficiency [162]. NaFeS₂ also shows an adsorption capacity of up to 129.3 mg g^{−1} for

methylene blue (MB) and can rapidly photodegrade 98% of MB within 20 min. This efficiency is 67 times that of TiO_2 .

The recycling of S-LFP cathode materials can extend beyond the recovery of lithium resources and the preparation of R-LFP. Furthermore, leveraging the inherent characteristics of S-LFP, such as lithium vacancies and uniform particle size, it can be converted into various functional materials with high economic value and other post-metal-ion battery components. However, there are common issues including complex conversion steps and excessively long processes. The resulting problems, such as high process treatment costs and poor product consistency, require further research to address.

In Table 3, the principal parameters that govern regeneration performance are collated, such as reaction temperature, ball milling rotation speed, reaction time, and reagent cost. It should be noted that the specific process of the optimized regeneration method is determined by the characteristics of the target material field. The optimal process should not only ensure the high value of the resulting material but also guarantee that the technical route is green and economical.

This section discusses in detail the application of upcycling method in recycling S-LFP. Unlike simple performance restoration, upcycling method focuses on component upgrading. This section classifies it is based on structural optimization, phase transformation, and conversion into other functional materials. Structural optimization improves the electrochemical performance of LFP by introducing element-doped carbon coatings. The selection of appropriate types and dosages of doping elements is crucial for structural optimization. To further enhance the energy density of LFP, converting LFP into high-performance LFMP through phase transformation (by introducing Mn) is also a current research focus. However, this process requires precise control over the uniform distribution of Fe/Mn and the appropriate content of Mn. The two aforementioned methods are limited to LIBs manufacturing. To further increase the economic value of products, converting S-LFP into post-metal-ion batteries and catalytic materials has become another research hotspot. Nevertheless, this conversion route generally faces issues such as complex steps and high costs, which require further research to resolve.

5 | Conclusion and Perspective

This review traces the root causes of LFP failure and discusses the characterization methods for analyzing the failure mechanism. With the help of advanced characterization techniques, it intuitively demonstrates the changes in the microscopic morphology of LFP during battery cycling, the phase transformation process, and the transitions in its structure and valence states. The failure analysis of spent batteries provides a research foundation and feasibility assessment for the upcycling of LFP (Figure 7A). Subsequently, the principles and methods of hydrometallurgy, direct regeneration, and upcycling are reviewed in detail. All of these methods are effective means to reduce environmental pollution, promote resource recycling, and advance the sustainable development of energy. However, hydrometallurgy inevitably requires large amounts of chemical reagents for leaching, coupled with its complex process flows and high energy consumption, which has gradually made it fall

out of favor. In contrast, direct regeneration aims to restore S-LFP materials to their original state with lower energy consumption and a simpler process, which is in line with the concept of green development. What is more advanced is that upcycling technology has gained favor among researchers due to its high-value transformation through structural reconstruction and functional upgrading, enabling efficient resource utilization. This provides a new direction for the recycling of S-LFP that combines environmental benefits with economic value.

It is worth noting that most current direct regeneration methods are only applicable to single cathode materials and cannot achieve simultaneous recycling of mixed cathode materials. In contrast, upcycling methods can simultaneously recycle S-LFP and S-LMO, converting them into high voltage and more stable LFMP to meet the demand for LIB with higher energy density in practical applications. Additionally, the modification of S-LFP through doping and coating can also achieve a comprehensive improvement in the electrochemical performance of cathode materials, providing new ideas for green and efficient upcycling methods of S-LFP. Moreover, the structural defects of S-LFP may provide beneficial functions in other fields (such as SIB and catalytic materials). More importantly, the upcycling process depends on specific application fields. The upcycling method takes high-value-added products as its core orientation, breaking through the traditional element extraction model in the field of battery recycling and advancing toward the development stage of functional material manufacturing. To evaluate the effectiveness of a regeneration method, in addition to assessing whether the performance of the resulting material is excellent, it is also necessary to analyze whether its process steps are environmentally friendly and whether the economic benefits meet the expected results.

Researchers frequently employ the EverBatt model, developed by Argonne National Laboratory [163], to simulate diverse recycling and regeneration pathways and to benchmark their feasibility with respect to energy consumption and economic performance. The model systematically contrasts alternative recycling routes across five cardinal dimensions: (i) cost advantage, (ii) recyclability, (iii) profit advantage, (iv) greenhouse gas (GHG) mitigation potential, and (v) energy conservation, thereby delivering an integrated economic-environmental assessment. Among these metrics, profit advantage is dissected with the finest granularity, because economic profitability constitutes the principal driver for industrial-scale implementation. Profit advantage is decomposed into expenditure and revenue streams. The payout includes reagent costs, energy costs, capital depreciation of equipment, and environmental-compliance costs, whereas the revenue comprises the market value of recovered materials, including battery grade metals, steel or aluminum casings, copper and aluminum current collectors, and anodic graphite. Because the recovered active materials represent the dominant revenue flux, raising their added economic value is the most efficacious lever for enhancing overall income. Upcycling strategies that yield high-performance electrode or catalytic materials command substantially higher market prices than the metal salts produced via hydrometallurgical routes, thereby generating superior profit margins. However, mainstream current analyses do not incorporate fiscal outflows (taxation) and policy-based inflows (government subsidies) into profit accounting. To enhance the objectivity and practical relevance of the profit advantage metric,

TABLE 3 | Information on upcycling method of S-LFP.

Method	Reagent	Condition	Performance	Reference
Optimizing the structure	S-LFP + CH ₃ COOLi + PAN + Glucose	① 500 rpm 4 h, 500 rpm 2 h ② 180°C 2 h ③ 400°C 5 h, 700°C 10 h	155 mAh g ⁻¹ (0.1 C), 87% (400r, 1 C)	[139]
	Li ₂ CO ₃ + 2% SmN ₃ O ₉ ·H ₂ O	Plasma ball milling: 1200 rpm 30 min, 700°C 6 h	142.2 mAh g ⁻¹ , 96.5% (200r, 1 C)	[140]
	CuO and S-LFP (200ppm: 1) + Glucose + Li ₂ CO ₃	① CuO + S-LFP: 500°C ② 400 rpm 2 h, 350°C 4 h, 700°C 10 h	130 mAh g ⁻¹ , 100% (300r, 2 C)	[146]
Changing the phase	① S-LFP + 0.2 M LiOH + 0.08 M tea polyphenols + H ₂ O ② P-LFP + 2 wt% Li ₂ CO ₃ + 3 wt% Al(NO ₃) ₃ /NH ₄ H ₂ PO ₄	① 120°C 10 h ② 600°C 5 h	124.3 mAh g ⁻¹ , 92.1% (400r, 2 C)	[148]
	① S-LFP + S-LMO + ChCl + OA ② P-LFMO: NH ₄ H ₂ PO ₄ : LiOH = 1:1: 1.05	① DES: 80°C, DES + S-LFP, 110°C 6 h ② 350°C 6 h, 650°C 10 h	ICE: 94%(1 C), 559 Wh kg ⁻¹	[141]
	S-LFP: Li ₂ CO ₃ : MnCO ₃ : NH ₄ H ₂ PO ₄ : Glucose = 100: 12.5: 15.6: 5.2: 20 ① S-LFP/S-LMO + H ₂ SO ₄ + 3% H ₂ O ₂ ② Leachate + 1 wt%	400 rpm 24 h 700°C 5 h ① 60°C/40°C 2 h (S/ L = 10 ml/g) ② 180°C 6 h ③ 350°C 2 h, 650°C 5 h	563.5 mAh g ⁻¹ , 83% (900r, 0.5 C)	[142]
	AA + MnSO ₄ ·2H ₂ O + FeSO ₄ ·7H ₂ O + H ₃ PO ₄ + LiOH·H ₂ O ① S-LFP + 2.5 M H ₂ SO ₄ + AA/MnCO ₃ /NH ₃ H ₂ PO ₄ ② Liquid + Polyethylene glycol solution	① Dissolving: 80°C ② 180°C 12 h ③ 650°C 5 h ① 550°C Ultrasonic: 30 min ② 240°C 2 h	144.2 mAh g ⁻¹ , 87% (1000r, 1 C), 136.9 mAh g ⁻¹ , 86.4% (1000r, 5 C)	[153]
Other upcycling methods	S-LFP + Ni(acac) ₂ + ethanol	① 550°C Ultrasonic: 30 min ② 240°C 2 h	561 Wh kg ⁻¹ 83.8% (2500r, 5 C)	[154]
	① S-LFP + Na ₂ SO ₃ + H ₂ O ② FePO ₄ + Na ₄ P ₂ O ₇ + C ₆ H ₈ O ₇	① Ball milling for 12 h ② 600°C 12 h	Overpotential: 268 mV and 292 mV, corresponding to oxygen evolution currents of 10 and 100 mA cm ⁻²	[160]
	① S-LFP + 50 ml H ₂ O + Na ₂ S ₂ O ₈ ② Lithium free powder + H ₂ O + Na ₂ S	① 25°C 40 min ② Reaction 25°C	82.9% (4000 r, 10 C) Adsorption: 129.3 mg g ⁻¹ , photodegrading 98% MB within 20 min	[143] [144]

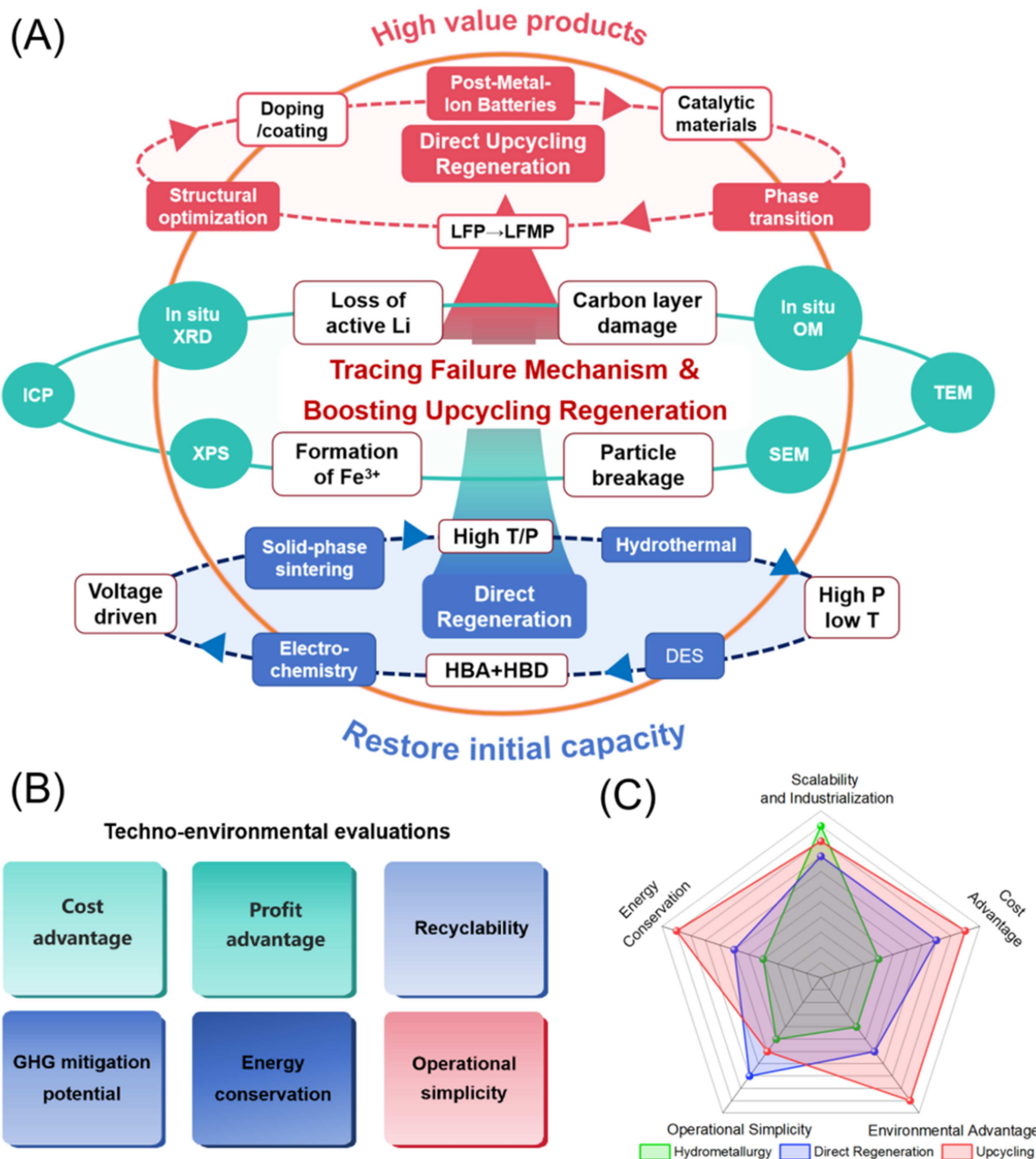


FIGURE 7 | (A) Schematic diagram summarizing the failure mechanism, direct regeneration, and upcycling of S-LFP, (B) The six primary dimensions of economic-environmental evaluation, (C) The Radar chart of hydrometallurgy, direct regeneration, and upcycling in five aspects.

both tax obligations and subsidy entitlements must be explicitly included in the accounting scope. Additionally, operational simplicity, an attribute often neglected in techno-environmental evaluations, has recently been proposed by Liu et al., [164] who quantified the comparative ease of disparate recycling flows. Operational simplicity subsumes four intertwined criteria: procedural straightforwardness, stringency of equipment specifications, environmental footprint, and requisite skill level of the workforce. These factors are essential for the promotion and healthy development of the industrial market for battery recycling (Figure 7B). Through the integrated economic-environmental assessment, it be concluded that direct regeneration/upcycling is the most environmentally friendly and achieves the highest profit return rate. However, they are still in the initial research stage. The comparison chart of hydrometallurgy, direct regeneration, and upcycling

in five aspects, including scalability and industrialization, energy conservation, and operational simplicity, is shown in Figure 7C. Thus, unavoidable challenges remain: how to simplify process steps, develop batteries or other functional materials with higher electrochemical performance, and scale up from laboratory research to industrial production. To address these difficulties and challenges, some perspectives are proposed from the following aspects:

1. Advanced in situ characterization techniques are employed to detect the damage status and composition information of spent cathode materials, laying a theoretical foundation for subsequent regeneration research. In situ and operando characterization techniques can real-time monitor the crystal structure distortion, particle

fragmentation degree, surface element loss, and composition changes of materials in the actual reaction environment. This breaks through the limitation of static sampling in traditional characterization and avoids the misleading of data deviation on regeneration schemes. Firstly, cathode materials with different failure degrees require different regeneration methods: slightly failed materials can restore performance through mild lithium supplementation, while severely failed ones need high-temperature treatment to reconstruct the crystal structure. In situ characterization techniques can real-time track the morphological (whether particles are recombined), structural (whether characteristic diffraction peaks are restored), and compositional changes of materials during the regeneration process. They reveal the key reaction steps and the causes of side reactions, helping researchers accurately match regeneration methods. This not only prevents resource waste due to over-treatment but also avoids substandard performance caused by under-treatment, thereby maximizing the residual value of materials. Moreover, mixed spent cathode materials available in the market include various LIB cathode materials (LCO, LFP, NMC, NCA). However, current regeneration methods are mostly applicable to single cathode materials. Direct treatment of mixed materials may easily lead to impurity generation due to incompatible processes, reducing the performance of regenerated materials. Preliminary screening via in situ characterization techniques can accurately identify the type, content, and failure degree of each component in mixed materials, enabling classified regeneration. By selecting suitable processes for materials with different components, resource utilization efficiency is significantly improved, promoting the large-scale and high-value recycling of mixed spent cathode materials.

2. Priority should be given to the upcycling method for treating S-LFP, so as to improve the economic value and performance of regenerated products. From the comparison of existing technologies in industry, hydrometallurgy can only recover valuable metals such as lithium and iron, and subsequent re-synthesis of battery materials is required before they can be put into use. This results in a long overall industrial chain and limited room for increasing added value. Direct regeneration can only restore the capacity of S-LFP. Although it shortens the treatment process, its functional positioning is still limited to LFP batteries. It cannot break through the constraints of original application scenarios and is difficult to meet the needs of high specific applications. In contrast, upcycling not only repairs the microstructural defects of materials but also converts S-LFP into other high performance functional materials by introducing new elements or adjusting the crystal phase structure. For example, it can be transformed into cathode materials for non-lithium metal ion batteries such as SIBs, which adapt to the current demands of the energy storage field for low cost, fast charging/discharging, high energy density, and other properties. It can also be converted into high-

efficiency photocatalysts for the degradation of organic pollutants or hydrogen production via water splitting, as well as OER catalysts in water electrolysis, contributing to the green development of the hydrogen energy industry chain. Upcycling expands the role of S-LFP from a single recycling target for battery materials to a precursor for diversified functional materials, with application scenarios covering multiple high-value fields such as new energy storage, environmental protection catalysis, and clean energy production. At the same time, since the converted functional materials usually have higher market added value, and the process flow features higher resource utilization efficiency and less secondary pollution, this method not only significantly improves the economic benefits of S-LFP treatment, but also provides key technical support for the industry to achieve circular development.

3. Machine learning (ML) methods should be integrated to predict the effectiveness of selected reagents and the performance of regenerated materials, enabling precision and efficiency throughout the entire regeneration process. To obtain optimal experimental conditions and regeneration performance, traditional approaches rely on conducting extensive experiments in laboratories. However, this method rarely achieves its goals due to the vast composition space, and it also consumes significant costs and time. In contrast, ML can leverage existing experimental data, such as molecular structures, doping systems, regeneration temperatures, and electrochemical performance, to construct multi-dimensional prediction models by combining different algorithms. These models enable rapid prediction of regenerated electrode performance, facilitating the identification of optimal experimental conditions. For example, Wang et al. [165] used an ML model to investigate how different types and concentrations of dopants affect the electrochemical performance of cathode materials, establishing a system for correlating doping characteristics with performance. By comparing key parameters of various models, Mohammed et al. [166] selected the model with the highest accuracy to predict the rate capability and cycle stability of regenerated materials under different experimental conditions, ultimately determining the optimal protocol. For lithium supplementation using chemical reagents, Tang et al. [167] employed an ML model to quickly predict the redox potential of different organic acids, their reaction activity with S-LFP, and lithium supplementation efficiency, leading to the development of a high-efficiency lithium supplementation scheme with optimized component ratios. These cases demonstrate the feasibility of ML and its broad prospects for application in optimizing regeneration processes. For example, ML can take parameters such as the atomic radius, electronegativity, and doping concentration of transition metals as inputs. However, ML lacks a comprehensive database dedicated to recycling processes, at present. Therefore, it calls for collaborative efforts among researchers to establish such a database, which could include detailed information on

leaching conditions and their corresponding outcomes, as well as lithium supplementation reagents and their effectiveness. More importantly, currently, most ML models rely predominantly on mathematical logic, which, while statistically valid, may not always align with physical principles or chemical realities, leading to inevitable invalid outputs. Therefore, physical and chemical logic should be integrated into ML models to enhance their consistency and impose meaningful constraints, thereby avoiding the common issue of lack of interpretability.

4. The goal is to establish a unified economic and environmental analysis system to clarify the economic and environmental feasibility of technical routes. Currently, some evaluation standards for regeneration methods only focus on the direct costs of the regeneration process (such as expenses for reagents and energy consumption), while ignoring the hidden costs throughout the entire material recycling lifecycle (such as costs for wastewater treatment and solid waste disposal). Additionally, some standards emphasize short-term economic benefits without accurately quantifying the potential costs of environmental impacts (such as carbon emissions and heavy metal pollution). Therefore, this system should be based on the entire lifecycle. From the economic dimension, it needs to consider factors such as the costs of different regeneration processes, fluctuations in market conditions, and policy changes. From the environmental dimension, it should quantify factors including energy consumption and greenhouse gas emissions of different regeneration processes. The combination of these two dimensions enables the coordinated evaluation of economic benefits and environmental impacts, ultimately providing standardized support for the formulation of technical routes. Through this standardized analysis framework, the current barrier of fragmented evaluation standards can be broken. It not only helps researchers clarify the direction of technical optimization but also provides enterprises with clear references for costs, benefits, and environmental impacts when selecting industrialization technical routes. Ultimately, it promotes the high-quality development of the S-LFP regeneration industry toward a direction that balances economic benefits and environmental sustainability.

Acknowledgements

This study was supported by the National Key Research and Development Program of China (No. 2022YFB2404300), the Science and Technology Special Fund of Hainan Province (No. ZDYF2025GXJS008) and the Hubei Provincial Science and Technology Innovation Talent Scheme Project (No. 2024DJA006).

Conflicts of Interest

The authors declare no conflicts of interest.

References

1. H. Zhao, H. D. Deng, A. E. Cohen, et al., "Learning Heterogeneous Reaction Kinetics From X-Ray Videos Pixel by Pixel," *Nature* 621, no. 7978 (2023): 289–294.

2. P. Li, S. Luo, Y. Lin, et al., "Fundamentals of the Recycling of Spent Lithium-Ion Batteries," *Chemical Society Reviews* 53, no. 24 (2024): 11967–12013.
3. J. Wang, J. Ma, Z. Zhuang, et al., "Toward Direct Regeneration of Spent Lithium-Ion Batteries: A Next-Generation Recycling Method," *Chemical Reviews* 124, no. 5 (2024): 2839–2887.
4. Y. Zhao, H. Du, Y. Kang, et al., "Spent Battery Regeneration for Better Recycling," *Nature Reviews Materials* 10 (2025): 722–724.
5. R. Fang, K. Chen, Z. Sun, G. Hu, D. W. Wang, and F. Li, "Realizing High-Energy Density for Practical Lithium-Sulfur Batteries," *Interdisciplinary Materials* 2, no. 5 (2023): 761–770.
6. X. Sun, Y. Zhou, D. Li, et al., "A Review of Electrospun Separators for Lithium-Based Batteries: Progress and Application Prospects," *Carbon Energy* 6, no. 9 (2024): e539.
7. J. J. Roy, D. M. Phuong, V. Verma, et al., "Direct Recycling of Li-Ion Batteries From Cell to Pack Level: Challenges and Prospects on Technology, Scalability, Sustainability, and Economics," *Carbon Energy* 6, no. 6 (2024): e492.
8. Lithium-Ion Battery Market Size, Share, and Trends 2025 to 2034, accessed May 28, 2025, <https://www.precedenceresearch.com/lithium-ion-battery-recyclingmarket>.
9. E. Fan, L. Li, Z. Wang, et al., "Sustainable Recycling Technology for Li-Ion Batteries and Beyond: Challenges and Future Prospects," *Chemical Reviews* 120, no. 14 (2020): 7020–7063.
10. TrendForce. Battery Recycling to Aid in Reducing Carbon Emissions, Global EV, Energy Storage Battery Recycling Market Forecast to Exceed 1TWh in 2030, accessed October 24, 2022, <https://www.trendforce.com/presscenter/news/20221024-11436.html>.
11. J. Yan, J. Qian, Y. Li, L. Li, F. Wu, and R. Chen, "Toward Sustainable Lithium Iron Phosphate in Lithium-Ion Batteries: Regeneration Strategies and Their Challenges," *Advanced Functional Materials* 34, no. 44 (2024): 2405055.
12. C. Qi, T. Yao, W. Zhai, M. Zhang, L. Song, and J. He, "Advances in Degradation Mechanism and Sustainable Recycling of LiFePO₄-Type Lithium-Ion Batteries," *Energy Storage Materials* 71 (2024): 103623.
13. M. Zhou, B. Li, J. Li, and Z. Xu, "Pyrometallurgical Technology in the Recycling of a Spent Lithium Ion Battery: Evolution and the Challenge," *ACS ES&T Engineering* 1, no. 10 (2021): 1369–1382.
14. J. C. Y. Jung, P. C. Sui, and J. Zhang, "A Review of Recycling Spent Lithium-Ion Battery Cathode Materials Using Hydrometallurgical Treatments," *Journal of Energy Storage* 35 (2021): 102217.
15. Z. J. Baum, R. E. Bird, X. Yu, and J. Ma, "Lithium-Ion Battery Recycling-Overview of Techniques and Trends," *ACS Energy Letters* 7, no. 2 (2022): 712–719.
16. T. Zhao, Y. Choi, C. Wu, et al., "A Review on Direct Regeneration of Spent Lithium Iron Phosphate: From Waste to Wealth," *Science of the Total Environment* 957 (2024): 177748.
17. X. Liu, R. Wang, S. Liu, et al., "Organic Eutectic Salts-Assisted Direct Lithium Regeneration for Extremely Low State of Health Ni-Rich Cathodes," *Advanced Energy Materials* 13, no. 44 (2023): 2302987.
18. X. Liu, S. Liu, J. Pu, et al., "Sustainable and Direct Upcycling of Waste Graphite Anodes via Deep Eutectic Solvents," *Advanced Science* 12 (2025): e06637.
19. R. Fang, K. Chen, Z. Sun, G. Hu, D. W. Wang, and F. Li, "Realizing High-Energy Density for Practical Lithium-Sulfur Batteries," *Interdisciplinary Materials* 2, no. 5 (2023): 761–770.
20. H. Ji, J. Wang, X. Qiu, et al., "A Universal Protocol for Ultrafast Direct Regeneration and Upcycling of Spent Lithium-Ion Battery Cathode Materials," *Nature Protocols* 36, no. 36 (2025): 2407029.
21. J. Wang, H. Ji, J. Li, et al., "Direct Recycling of Spent Cathode Material at Ambient Conditions via Spontaneous Lithiation," *Nature Sustainability* 7 (2024): 1283–1293.

22. W. Chen, J. Li, H. Ji, et al., "Efficient and Scalable Direct Regeneration of Spent Layered Cathode Materials via Advanced Oxidation," *Advanced Materials* 37, no. 9 (2025): 2416818.
23. H. Ji, J. Wang, H. Qu, et al., "Closed-Loop Direct Upcycling of Spent Ni-Rich Layered Cathodes Into High-Voltage Cathode Materials," *Advanced Materials* 36, no. 36 (2024): 2407029.
24. W. Wang, R. Wang, R. Zhan, et al., "Probing Hybrid LiFePO₄/FePO₄ Phases in a Single Olive LiFePO₄ Particle and Their Recovering From Degraded Electric Vehicle Batteries," *Nano Letters* 23, no. 16 (2023): 7485–7492.
25. M. Zheng, Y. You, and J. Lu, "Understanding Materials Failure Mechanisms for the Optimization of Lithium-Ion Battery Recycling," *Nature Reviews Materials* 10 (2025): 355–368.
26. Z. Qin, Y. Zhang, W. Luo, et al., "A Universal Molten Salt Method for Direct Upcycling of Spent Ni-Rich Cathode Towards Single-Crystalline Li-Rich Cathode," *Angewandte Chemie International Edition* 62, no. 25 (2023): e202218672.
27. C. R. Birkel, M. R. Roberts, E. McTurk, P. G. Bruce, and D. A. Howey, "Degradation Diagnostics for Lithium Ion Cells," *Journal of Power Sources* 341 (2017): 373–386.
28. L. Guo, Y. Zhang, J. Wang, et al., "Unlocking the Energy Capabilities of Micron-Sized LiFePO₄," *Nature Communications* 6, no. 1 (2015): 7898.
29. A. Andersson, "Lithium Extraction/Insertion in LiFePO₄: An X-Ray Diffraction and Mössbauer Spectroscopy Study," *Solid State Ionics* 130, no. 1–2 (2000): 41–52.
30. M. S. Islam, D. J. Driscoll, C. A. J. Fisher, and P. R. Slater, "Atomic-Scale Investigation of Defects, Dopants, and Lithium Transport in the LiFePO₄ Olivine-Type Battery Material," *Chemistry of Materials* 17, no. 20 (2005): 5085–5092.
31. H. Ji, J. Wang, J. Ma, H. M. Cheng, and G. Zhou, "Fundamentals, Status and Challenges of Direct Recycling Technologies for Lithium Ion Batteries," *Chemical Society Reviews* 52, no. 23 (2023): 8194–8244.
32. C. Ouyang, S. Shi, Z. Wang, X. Huang, and L. Chen, "First-Principles Study of Li Ion Diffusion in LiFePO₄," *Physical Review B* 69, no. 10 (2004): 104303.
33. L. Hong, L. Li, Y. K. Chen-Wiegar, et al., "Two-Dimensional Lithium Diffusion Behavior and Probable Hybrid Phase Transformation Kinetics in Olivine Lithium Iron Phosphate," *Nature Communications* 8, no. 1 (2017): 1194.
34. Z. Zeng, H. Lei, X. Lu, et al., "Li-Fe Anti-Sites Defects in LiFePO₄: Mechanism, Characterization and Cathode-Regeneration Applications," *Energy Storage Materials* 74 (2024): 103947.
35. R. Malik, D. Burch, M. Bazant, and G. Ceder, "Particle Size Dependence of the Ionic Diffusivity," *Nano Letters* 10, no. 10 (2010): 4123–4127.
36. X. Lv, J. Lin, X. Sun, et al., "Direct Recycling of Spent LiFePO₄ Cathodes Through Photocatalytic Correction of Anti-Site Defects," *Advanced Materials* 37, no. 26 (2025): 2503398.
37. H. Zheng, L. Tan, L. Zhang, et al., "Correlation Between Lithium Deposition on Graphite Electrode and the Capacity Loss for LiFePO₄/Graphite Cells," *Electrochimica Acta* 173 (2015): 323–330.
38. Y. L. Han, H. Wang, H. F. Di, et al., "Determination of High-Temperature Float Charge Failure Mechanisms in Lithium-Ion Batteries by Quantifying Active Lithium Loss," *Carbon Energy* 7, no. 7 (2025): e70002.
39. Y. Kim, Y. Kim, S. Yeo, et al., "Development of a Continuous Hydrogen Generator Fueled by Ammonia Borane for Portable Fuel Cell Applications," *Journal of Power Sources* 229 (2013): 170–178.
40. J. Wu, R. A. Davies, M. S. Islam, and S. M. Hailu, "Atomistic Study of Doped BaCeO₃: Dopant Site-Selectivity and Cation Non-stoichiometry," *Chemistry of Materials* 17, no. 4 (2005): 846–851.
41. L. Wei and C. Shifu, "Preparation and Characterization of p-n Heterojunction Photocatalyst Cu₂O/In₂O₃ and Its Photocatalytic Activity under Visible and UV Light Irradiation," *Journal of the Electrochemical Society* 157, no. 11 (2010): H1029.
42. X. Han, L. Lu, Y. Zheng, et al., "A Review on the Key Issues of the Lithium Ion Battery Degradation Among the Whole Life Cycle," *eTransportation* 1 (2019): 100005.
43. Z. Ren, H. Li, Y. Wang, et al., "Green Recycling Assessment on Typical Spent Lithium-Ion Batteries (LIBs): A Multi-Objective Assessment," *Resources, Conservation And Recycling* 206 (2024): 107648.
44. D. Wang, X. Wu, Z. Wang, and L. Chen, "Cracking Causing Cyclic Instability of LiFePO₄ Cathode Material," *Journal of Power Sources* 140, no. 1 (2005): 125–128.
45. A. K. Padhi, K. S. Nanjundaswamy, and J. B. Goodenough, "Phospho-Olivines as Positive-Electrode Materials for Rechargeable Lithium Batteries," *Journal of the Electrochemical Society* 144, no. 4 (1997): 1188–1194.
46. D. Morgan, A. Van der Ven, and G. Ceder, "Li Conductivity in Li_xMPO₄ (M = Mn, Fe, Co, Ni) Olivine Materials," *Electrochemical and Solid-State Letters* 7, no. 2 (2004): A30.
47. S. Kim, H. R. Shin, K. J. Kim, M. S. Park, and J. W. Lee, "Driving Inward Growth of Lithium Metal in Hollow Microcapsule Hosts by Heteroatom-Controlled Nucleation," *Carbon Energy* 6, no. 8 (2024): e525.
48. D. Peng, X. Wang, S. Wang, et al., "Efficient Regeneration of Retired LiFePO₄ Cathode by Combining Spontaneous and Electrically Driven Processes," *Green Chemistry* 24, no. 11 (2022): 4544–4556.
49. H. Zhang, Y. Yang, D. Ren, L. Wang, and X. He, "Graphite as Anode Materials: Fundamental Mechanism, Recent Progress and Advances," *Energy Storage Materials* 36 (2021): 147–170.
50. P. Verma, P. Maire, and P. Novák, "A Review of the Features and Analyses of the Solid Electrolyte Interphase in Li-Ion Batteries," *Electrochimica Acta* 55, no. 22 (2010): 6332–6341.
51. D. Aurbach, E. Zinigrad, Y. Cohen, and T. Hanan, "A Short Review of Failure Mechanisms of Lithium Metal and Lithiated Graphite Anodes in Liquid Electrolyte Solutions," *Solid State Ionics* 148, no. 3–4 (2002): 405–416.
52. M. Li, S. Yang, and B. Li, "Advances in Electrolyte-Anode Interface Engineering of Solid-State Lithium Metal Batteries," *Interdisciplinary Materials* 3, no. 6 (2024): 805–834.
53. N. Zhu, Y. Yang, Y. Li, Y. Bai, J. Rong, and C. Wu, "Carbon-Based Interface Engineering and Architecture Design for High-Performance Lithium Metal Anodes," *Carbon Energy* 6, no. 1 (2024): e423.
54. P. Ding, Z. Lin, X. Guo, et al., "Polymer Electrolytes and Interfaces in Solid-State Lithium Metal Batteries," *Materials Today* 51 (2021): 449–474.
55. D. Ouyang, M. Chen, J. Liu, R. Wei, J. Weng, and J. Wang, "Investigation of A Commercial Lithium-Ion Battery Under Overcharge/Over-Discharge Failure Conditions," *RSC Advances* 8, no. 58 (2018): 33414–33424.
56. M. Qin, Z. Zeng, S. Cheng, and J. Xie, "Challenges and Strategies of Formulating Low-Temperature Electrolytes in Lithium-Ion Battery," *Interdisciplinary Materials* 2, no. 2 (2023): 308–336.
57. S. Grolleau, A. Delaille, H. Gualous, et al., "Calendar Aging Of Commercial Graphite/LiFePO₄ Cell-Predicting Capacity Fade Under Time Dependent Storage Conditions," *Journal of Power Sources* 255 (2014): 450–458.
58. M. Wang, K. Liu, S. Dutta, et al., "Recycling of Lithium Iron Phosphate Batteries: Status, Technologies, Challenges, and Prospects," *Renewable and Sustainable Energy Reviews* 163 (2022): 112515.
59. M. K. Tufail, P. Zhai, W. Khokar, M. Jia, N. Zhao, and X. Guo, "Evaluation of Solid Electrolytes: Development of Conventional and

- Interdisciplinary Approaches," *Interdisciplinary Materials* 2, no. 4 (2023): 529–568.
60. L. Bertoluzzi, C. C. Boyd, N. Rolston, et al., "Mobile Ion Concentration Measurement and Open-Access Band Diagram Simulation Platform for Halide Perovskite Solar Cells," *Joule* 4, no. 1 (2020): 109–127.
61. J. Tang, H. Qu, C. Sun, et al., "A Universal Solution for Direct Regeneration of Spent Lithium Iron Phosphate," *Advanced Materials* 37, no. 14 (2025): 2420238.
62. C. Hogrefe, T. Waldmann, M. Hölzle, and M. Wohlfahrt-Mehrens, "Direct Observation of Internal Short Circuits by Lithium Dendrites in Cross-Sectional Lithium-Ion in Situ Full Cells," *Journal of Power Sources* 556 (2023): 232391.
63. Y. Lin, H. Liu, C. Wang, et al., "Transient Atom-Economy Utilization of Residual Binder for Upcycling the Spent LiFePO₄ Cathodes," *CCS Chemistry* 7 (2025): e202506030.
64. K. Jia, J. Ma, J. Wang, et al., "Long-Life Regenerated LiFePO₄ From Spent Cathode by Elevating the d-Band Center of Fe," *Advanced Materials* 35, no. 5 (2023): 2208034.
65. X. Huang, X. He, C. Jiang, G. Tian, and Y. Liu, "Reaction Mechanisms on Solvothermal Synthesis of Nano LiFePO₄ Crystals and Defect Analysis," *Industrial & Engineering Chemistry Research* 56, no. 38 (2017): 10648–10657.
66. U. Boesenberg, F. Meirer, Y. Liu, et al., "Mesoscale Phase Distribution in Single Particles of LiFePO₄ Following Lithium Deintercalation," *Chemistry of Materials* 25, no. 9 (2013): 1664–1672.
67. N. Meethong, H. Y. S. Huang, S. A. Speakman, W. C. Carter, and Y. M. Chiang, "Strain Accommodation During Phase Transformations in Olivine-Based Cathodes as a Materials Selection Criterion for High-Power Rechargeable Batteries," *Advanced Functional Materials* 17, no. 7 (2007): 1115–1123.
68. L. Li, Y. Bian, X. Zhang, et al., "A Green and Effective Room-Temperature Recycling Process of LiFePO₄ Cathode Materials for Lithium-Ion Batteries," *Waste Management* 85 (2019): 437–444.
69. Z. Liang, C. Cai, G. Peng, et al., "Hydrometallurgical Recovery of Spent Lithium Ion Batteries: Environmental Strategies and Sustainability Evaluation," *ACS Sustainable Chemistry & Engineering* 9, no. 17 (2021): 5750–5767.
70. P. Yadav, C. J. Jie, S. Tan, and M. Srinivasan, "Recycling of Cathode From Spent Lithium Iron Phosphate Batteries," *Journal of Hazardous Materials* 399 (2020): 123068.
71. J. J. Roy, S. Rarotra, V. Krikstolaityte, et al., "Green Recycling Methods to Treat Lithium-Ion Batteries E-Waste: A Circular Approach to Sustainability," *Advanced Materials* 34, no. 25 (2022): 2103346.
72. Y. Yang, E. G. Okonkwo, G. Huang, S. Xu, W. Sun, and Y. He, "On the Sustainability of Lithium Ion Battery Industry-A Review and Perspective," *Energy Storage Materials* 36 (2021): 186–212.
73. T. Zhao, W. Li, M. Traversy, et al., "A Review on the Recycling of Spent Lithium Iron Phosphate Batteries," *Journal of Environmental Management* 351 (2024): 119670.
74. Y. Xu, B. Zhang, Z. Ge, et al., "Advances and Perspectives Towards Spent LiFePO₄ Battery Recycling," *Journal of Cleaner Production* 434 (2024): 140077.
75. M. Joulié, R. Laucournet, and E. Billy, "Hydrometallurgical Process for the Recovery of High Value Metals From Spent Lithium Nickel Cobalt Aluminum Oxide Based Lithium-Ion Batteries," *Journal of Power Sources* 247 (2014): 551–555.
76. Y. Yao, M. Zhu, Z. Zhao, B. Tong, Y. Fan, and Z. Hua, "Hydrometallurgical Processes for Recycling Spent Lithium-Ion Batteries: A Critical Review," *ACS Sustainable Chemistry & Engineering* 6, no. 11 (2018): 13611–13627.
77. J. Cong, S. Luo, Q. Sun, et al., "Study on Deep Synergistic Leaching Mechanism of Spent Lithium Iron Phosphate Under the H₂SO₄–H₂O₂ System and Precise Chemical Precipitation Recovery Strategy of Lithium Carbonate," *Separation and Purification Technology* 363 (2025): 132326.
78. X. Zhou, W. Yang, X. Liu, et al., "One-Step Selective Separation and Efficient Recovery of Valuable Metals From Mixed Spent Lithium Batteries in the Phosphoric Acid System," *Waste Management* 155 (2023): 53–64.
79. J. Kumar, X. Shen, B. Li, H. Liu, and J. Zhao, "Selective Recovery of Li and FePO₄ From Spent LiFePO₄ Cathode Scraps by Organic Acids and The Properties of the Regenerated LiFePO₄," *Waste Management* 113 (2020): 32–40.
80. D. F. Wang, M. Chen, J. J. Zhao, et al., "Revealing Role of Oxidation in Recycling Spent Lithium Iron Phosphate Through Acid Leaching," *Rare Metals* 44, no. 3 (2025): 2059–2070.
81. Y. Zhang, B. Wang, F. Wang, et al., "A Green Recyclable Process for Selective Recovery of Li and Fe From Spent Lithium Iron Phosphate Batteries by Synergistic Effect of Deep Eutectic Solvent and Oxygen," *Separation and Purification Technology* 354 (2025): 128764.
82. X. Li, M. Benstead, N. Peeters, and K. Binnemans, "Recycling of Metals From LiFePO₄ Battery Cathode Material by Using Ionic Liquid Based-Aqueous Biphasic Systems," *RSC Advances* 14, no. 13 (2024): 9262–9272.
83. W. Gao, X. Zhang, X. Zheng, et al., "Lithium Carbonate Recovery From Cathode Scrap of Spent Lithium-Ion Battery: A Closed-Loop Process," *Environmental Science & Technology* 51, no. 3 (2017): 1662–1669.
84. Y. Yang, X. Meng, H. Cao, et al., "Selective Recovery of Lithium From Spent Lithium Iron Phosphate Batteries: A Sustainable Process," *Green Chemistry* 20, no. 13 (2018): 3121–3133.
85. Q. Zeng, K. Hu, W. Hu, et al., "Suppressing the Coordination of Ferric Ions for Efficient Recycling of Spent Lithium Iron Phosphate," *Journal of Environmental Chemical Engineering* 13, no. 3 (2025): 117060.
86. E. Colacino, V. Isoni, D. Crawford, and F. García, "Upscaling Mechanochemistry: Challenges and Opportunities for Sustainable Industry," *Trends in Chemistry* 3, no. 5 (2021): 335–339.
87. J. Neumann, M. Petranikova, M. Meeus, et al., "Recycling of Lithium-Ion Batteries-Current State of the Art, Circular Economy, and Next Generation Recycling," *Advanced Energy Materials* 12, no. 17 (2022): 2102917.
88. L. Takacs, "The Historical Development of Mechanochemistry," *Chemical Society Reviews* 42, no. 18 (2013): 7649–7659.
89. Q. Zhang, E. Fan, J. Lin, et al., "Acid-Free Mechanochemical Process to Enhance the Selective Recycling of Spent LiFePO₄ Batteries," *Journal of Hazardous Materials* 443 (2023): 130160.
90. Y. Jiang, X. Chen, S. Yan, Y. Ou, and T. Zhou, "Mechanochemistry-Induced Recycling of Spent Lithium-Ion Batteries for Synergistic Treatment of Mixed Cathode Powders," *Green Chemistry* 24, no. 15 (2022): 5987–5997.
91. Y. Hua, Z. Xu, B. Zhao, and Z. Zhang, "Electric Potential-Determined Redox Intermediates for Effective Recycling of Spent Lithium-Ion Batteries," *Green Chemistry* 24, no. 9 (2022): 3723–3735.
92. X. Ren, X. Hu, B. Zhang, et al., "Dual Contribution of Recyclable Oxidants and Generated Superoxide Anion (O₂^{•−}) for the Selective Recycling of Spent LiFePO₄ Batteries," *Separation and Purification Technology* 365 (2025): 132685.
93. J. Du, J. Qing, K. Fang, et al., "Efficient Lithium Recovery From Spent LiFePO₄ Cathodes via Alkaline Pressure Leaching," *Separation and Purification Technology* 378 (2025): 134700.
94. K. Liu, S. Yang, F. Lai, et al., "Application of H₄P₂O₇ as Leaching Acid in One-Step Selective Recovery for Metals From Spent LiFePO₄ Batteries," *Ionics* 27, no. 12 (2021): 5127–5135.

95. X. Qiu, B. Zhang, Y. Xu, et al., "Enabling the Sustainable Recycling of LiFePO_4 From Spent Lithium-Ion Batteries," *Green Chemistry* 24, no. 6 (2022): 2506–2515.
96. Y. Fan, Y. Kong, P. Jiang, et al., "Development and Challenges of Deep Eutectic Solvents for Cathode Recycling of End-of-Life Lithium-Ion Batteries," *Chemical Engineering Journal* 463 (2023): 142278.
97. K. Zhou, X. Dai, P. Li, et al., "Recent Advances in Deep Eutectic Solvents for Next-Generation Lithium Batteries: Safer and Greener," *Progress in Materials Science* 146 (2024): 101338.
98. C. Wang, R. Tao, P. Xing, et al., "State-of-the-Art Review on Liberation of Cathode Material in Pretreatment Recycling of Spent Lithium-Ion Battery," *Resources, Conservation And Recycling* 218 (2025): 108257.
99. J. Zou, R. Zhang, Y. Huang, et al., "High Efficiency Leaching of Black Powder From Spent Lithium-Ion Battery by Ternary Deep Eutectic Solvent and Recovery of Metals by Precipitation and Electrodeposition," *Separation and Purification Technology* 364 (2025): 132438.
100. M. Chen, X. Ma, B. Chen, et al., "Recycling End-of-Life Electric Vehicle Lithium-Ion Batteries," *Joule* 3, no. 11 (2019): 2622–2646.
101. H. Zhou, Z. Luo, S. Wang, X. Ma, and Z. Cao, "A Mild Closed-Loop Process for Lithium-Iron Separation and Cathode Materials Regeneration From Spent LiFePO_4 Batteries," *Separation and Purification Technology* 315 (2023): 123742.
102. X. Chen, S. Li, Y. Wang, et al., "Recycling of LiFePO_4 Cathode Materials From Spent Lithium-Ion Batteries Through Ultrasound-Assisted Fenton Reaction and Lithium Compensation," *Waste Management* 136 (2021): 67–75.
103. H. Gao, D. Tran, and Z. Chen, "Seeking Direct Cathode Regeneration for More Efficient Lithium-Ion Battery Recycling," *Current Opinion In Electrochemistry* 31 (2022): 100875.
104. Y. Wang, Y. Yang, J. Zhang, Y. Chen, and C. Wang, "Residue Carbon Removal for the High-Quality and Sustainable Direct Regeneration of Spent LiFePO_4 Materials," *Applied Surface Science* 689 (2025): 162512.
105. H. Xiao, C. Zeng, F. Fan, et al., "A One-Step Low-Temperature Closed-Loop Eutectic Salt Strategy for Direct Regeneration of Severely Degraded LiFePO_4 ," *Energy Storage Materials* 77 (2025): 104183.
106. G. Ji, J. Wang, Z. Liang, et al., "Direct Regeneration of Degraded Lithium-Ion Battery Cathodes With a Multifunctional Organic Lithium Salt," *Nature Communications* 14, no. 1 (2023): 584.
107. D. Tang, G. Ji, J. Wang, et al., "A Multifunctional Amino Acid Enables Direct Recycling of Spent LiFePO_4 Cathode Material," *Advanced Materials* 36, no. 5 (2024): 2309722.
108. Y. Yang, J. Zhang, H. Zhang, Y. Wang, Y. Chen, and C. Wang, "Simultaneous Anodic De-Lithiation/Cathodic Lithium-Embedded Regeneration Method for Recycling of Spent LiFePO_4 Battery," *Energy Storage Materials* 65 (2024): 103081.
109. S. Zhou, J. Du, X. Xiong, et al., "Direct Recovery of Scrapped LiFePO_4 by a Green and Low-Cost Electrochemical Re-Lithiation Method," *Green Chemistry* 24, no. 16 (2022): 6278–6286.
110. Y. Lin, T. Wang, C. Gao, et al., "Direct and Low-Temperature Regeneration of Degraded LiFePO_4 Cathodes at Ambient Conditions Using Green and Sustainable Deep Eutectic Solvent," *Advanced Science* 12, no. 8 (2025): 2504683.
111. P. Xu, Q. Dai, H. Gao, et al., "Efficient Direct Recycling of Lithium-Ion Battery Cathodes by Targeted Healing," *Joule* 4, no. 12 (2020): 2609–2626.
112. J. Sun, Z. Jiang, P. Jia, et al., "A Sustainable Revival Process for Defective LiFePO_4 Cathodes Through the Synergy of Defect-Targeted Healing and In-Situ Construction of 3D-Interconnected Porous Carbon Networks," *Waste Management* 158 (2023): 125–135.
113. C. Cheng, X. Cao, Z. Xing, and S. Tang, "Direct Regeneration of Severely Damaged Spent LiFePO_4 Cathodes," *Journal of Materials Science & Technology* 241, no. 10 (2026): 262–269.
114. Q. Jing, J. Zhang, Y. Liu, et al., "Direct Regeneration of Spent LiFePO_4 Cathode Material by a Green and Efficient One-Step Hydrothermal Method," *ACS Sustainable Chemistry & Engineering* 48, no. 48 (2020): 17622–17628.
115. K. Y. Park, I. Park, H. Kim, et al., "Anti-Site Reordering in LiFePO_4 : Defect Annihilation on Charge Carrier Injection," *Chemistry of Materials* 26, no. 18 (2014): 5345–5351.
116. J. Yan, J. Qian, Y. Li, L. Li, F. Wu, and R. Chen, "Toward Sustainable Lithium Iron Phosphate in Lithium-Ion Batteries: Regeneration Strategies and Their Challenges," *Advanced Functional Materials* 34, no. 44 (2024): 2405055.
117. T. Yang, D. Luo, A. Yu, and Z. Chen, "Enabling Future Closed-Loop Recycling of Spent Lithium-Ion Batteries: Direct Cathode Regeneration," *Advanced Materials* 35, no. 36 (2023): 2203218.
118. U. Nisar, N. Muralidharan, R. Essehli, R. Amin, and I. Belharouak, "Valuation of Surface Coatings in High-Energy Density Lithium-Ion Battery Cathode Materials," *Energy Storage Materials* 38 (2021): 309–328.
119. C. Feng, Y. Cao, L. Song, et al., "Direct Regeneration of Industrial LiFePO_4 Black Mass Through a Glycerol-Enabled Granule Reconstruction Strategy," *Angewandte Chemie International Edition* 64, no. 6 (2025): e202418198.
120. Z. Jiang, Z. Xu, L. Li, et al., "Design Principles for Efficient Hydrothermal Relithiation of Spent Lithium Iron Phosphate," *ACS Applied Materials & Interfaces* 17, no. 3 (2025): 4875–4883.
121. Y. Yang, Z. Liu, J. Zhang, Y. Chen, and C. Wang, "Economical and Low-Carbon Regeneration of Spent LiFePO_4 Materials by Hydrothermal Relithiation," *Journal of Alloys and Compounds* 947 (2023): 169660.
122. Z. Chi, J. Li, L. Wang, et al., "Direct Regeneration Method of Spent $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ Cathode Materials via Surface Lithium Residues," *Green Chemistry* 23, no. 22 (2021): 9099–9108.
123. S. Yang, H. Luo, Y. Li, et al., "Lithium Resurrection: Synergistic Thermal-Decomposition and Electric-Drive Strategy Enabling Inactive Lithium Fully Recycling," *Journal of Energy Chemistry* 102 (2025): 842–851.
124. P. Xu, D. H. S. Tan, B. Jiao, H. Gao, X. Yu, and Z. Chen, "A Materials Perspective on Direct Recycling of Lithium-Ion Batteries: Principles, Challenges and Opportunities," *Advanced Functional Materials* 33, no. 14 (2023): 2213168.
125. M. Fan, Q. Meng, X. Chang, et al., "In Situ Electrochemical Regeneration of Degraded LiFePO_4 Electrode With Functionalized Prelithiation Separator," *Advanced Energy Materials* 12, no. 18 (2022): 2103630.
126. T. El Achkar, H. Greige-Gerges, and S. Fourmentin, "Basics and Properties of Deep Eutectic Solvents: A Review," *Environmental Chemistry Letters* 19, no. 4 (2021): 3397–3408.
127. L. Chen, Y. Chao, X. Li, et al., "Engineering a Tandem Leaching System for the Highly Selective Recycling of Valuable Metals From Spent Li-Ion Batteries," *Green Chemistry* 23, no. 5 (2021): 2177–2184.
128. M. K. Tran, M. T. F. Rodrigues, K. Kato, G. Babu, and P. M. Ajayan, "Deep Eutectic Solvents for Cathode Recycling of Li-Ion Batteries," *Nature Energy* 4 (2019): 339–345.
129. S. Wang, Z. Zhang, Z. Lu, and Z. Xu, "A Novel Method for Screening Deep Eutectic Solvent to Recycle the Cathode of Li-Ion Batteries," *Green Chemistry* 22, no. 14 (2020): 4473–4482.
130. H. Pu, X. Zhu, Z. Zou, and C. Jiang, "Direct Regeneration of Spent LiFePO_4 Cathode via a Mild Deep Eutectic Solvent Process," *Journal of Energy Storage* 111 (2025): 115421.

131. T. Yingnakorn, J. Hartley, J. S. Terreblanche, C. Lei, W. M. Dose, and A. P. Abbott, "Direct Re-Lithiation Strategy for Spent Lithium Iron Phosphate Battery in Li-Based Eutectic Using Organic Reducing Agents," *RSC Sustainability* 1, no. 9 (2023): 2341–2349.
132. J. Shen, M. Zhou, W. Liu, et al., "Advanced Direct Recycling Technology Enables a Second Life of Spent Lithium-Ion Battery," *Energy Storage Materials* 74 (2024): 103964.
133. A. Khan, H. A. Rashid, P. K. Roy, et al., "Challenges and the Way to Improve Lithium-Ion Battery Technology for Next-Generation Energy Storage," *Energy & Environmental Materials* 8, no. 6 (2025): e70088.
134. L. Song, C. Qi, S. Wang, et al., "Direct Regeneration of Waste LiFePO₄ Cathode Materials With a Solid-Phase Method Promoted by Activated CNTs," *Waste Management* 157 (2023): 141–148.
135. X. Xu, Z. Guo, D. Zhu, J. Pan, C. Yang, and S. Li, "Green Pathways to LiFePO₄ Cathodes: Exploring Sustainable Iron Sources and Recycling Strategies," *Chemical Engineering Journal* 521 (2025): 166798.
136. Z. Zeng, P. Xu, J. Li, et al., "Large-Scale and Homogenized Strategies of Spent LiFePO₄ Recycling: Reconstruction of Targeted Lattice," *Advanced Functional Materials* 34, no. 6 (2024): 2308671.
137. S. Lei, W. Sun, and Y. Yang, "Comprehensive Technology for Recycling and Regenerating Materials From Spent Lithium Iron Phosphate Battery," *Environmental Science & Technology* 58, no. 8 (2024): 3609–3628.
138. L. Wang, H. Chen, Y. Zhang, J. Liu, and L. Peng, "Research Progress in Strategies for Enhancing the Conductivity and Conductive Mechanism of LiFePO₄ Cathode Materials," *Molecules* 29, no. 22 (2024): 5250.
139. M. Xiao, X. Fu, M. Chen, et al., "Constructing a Homogeneous Medium Layer to Promote the Direct Regeneration of Spent Lithium Iron Phosphate," *ACS Applied Materials & Interfaces* 17, no. 8 (2025): 12199–12207.
140. Y. Li, C. Li, F. Hu, et al., "Direct Regeneration of Spent LiFePO₄ Cathode Materials Through Li⁺ Supplementation and Sm Doping," *Nano Research Energy* 4, no. 4 (2025): e9120190.
141. G. Ji, D. Tang, J. Wang, et al., "Sustainable Upcycling of Mixed Spent Cathodes to a High-Voltage Polyanionic Cathode Material," *Nature Communications* 15, no. 4 (2024): 4086.
142. S. Ji, Y. Tan, J. Wang, et al., "Defect-Engineered Gradient Reconstruction for the Upcycling of Spent LiFePO₄ to Generate High-Value LiFe_{1-x}Mn_xPO₄/C Cathodes," *Journal of Energy Chemistry* 122 (2025): 306–316.
143. X. T. Wang, Z. Y. Gu, J. M. Cao, et al., "Dual-Loop Upcycling of Spent LiFePO₄: Defect Inheritance Enables Durable and Fast-Charging Sodium-Ion Batteries," *National Science Review* 12, no. 9 (2025): nwaf321.
144. X. H. Yue and F. S. Zhang, "Recycling Spent LiFePO₄ Battery for Fabricating Visible-Light Photocatalyst With Adsorption-Photocatalytic Synergistic Performance and Simultaneous Recovery of Lithium and Phosphorus," *Chemical Engineering Journal* 450, no. 4 (2022): 138388.
145. M. Zhang, M. Lv, D. Zhang, et al., "Enhanced Electrochemical Properties of NCM811 Cathode Material Due to Synergistic Modification With Sm as Doping and Coating Agent," *Journal of Alloys and Compounds* 909 (2022): 164712.
146. Z. Zeng, H. Lei, J. Li, et al., "Regenerated Spent LiFePO₄ With Tailored Residual Copper-Atoms Towards Improved Energy-Storage Capacity and Reversibility," *Chemical Engineering Journal* 499 (2024): 155616.
147. D. Wang, H. Li, S. Shi, X. Huang, and L. Chen, "Improving the Rate Performance of LiFePO₄ by Fe-Site Doping," *Electrochimica Acta* 50, no. 14 (2005): 2955–2958.
148. Y. Liu, J. Bai, R. Shi, et al., "Direct Recycling of Degraded LiFePO₄ Cathode Material via Natural Electron Donors Healing and Targeted Surface Reconstruction," *Advanced Materials* 38, no. 1 (2025): e11246.
149. Y. F. Meng, H. J. Liang, C. D. Zhao, et al., "Concurrent Recycling Chemistry for Cathode/Anode in Spent Graphite/LiFePO₄ Batteries: Designing A Unique Cation/Anion-Co-Workable Dual-Ion Battery," *Journal of Energy Chemistry* 64 (2022): 166–171.
150. V. Etacheri, R. Marom, R. Elazari, G. Salitra, and D. Aurbach, "Challenges in the Development of Advanced Li-Ion Batteries: A Review," *Energy & Environmental Science* 4, no. 9 (2011): 3243–3262.
151. T. J. Diethrich, S. Gnewuch, K. G. Dold, K. M. Taddei, and E. E. Rodriguez, "Tuning Magnetic Symmetry and Properties in the Olivine Series LiFe_{1-x}Mn_xPO₄ Through Selective Delithiation," *Chemistry of Materials* 34, no. 11 (2022): 5039–5053.
152. D. B. Ravnsbæk, K. Xiang, W. Xing, et al., "Extended Solid Solutions and Coherent Transformations in Nanoscale Olivine Cathodes," *Nano Letters* 14, no. 3 (2014): 1484–1491.
153. S. Lei, J. Li, W. Sun, P. Ge, and Y. Yang, "Upcycling of Low-Value Cathode Materials From Spent Lithium-Ion Battery to High-Voltage Cathode With Ultrahigh Rate Capability and Reversibility," *Advanced Energy Materials* 15, no. 21 (2025): 2406064.
154. C. Cheng, W. Mao, X. Cao, K. Xu, and S. Tang, "Sustainable Transformation of Spent Lifepo₄ Cathodes into High-Voltage Olivine LiMn_xFe_{1-x}PO₄ via Solvothermal Upcycling Strategy for Next-Generation Cathode Material," *Energy Storage Materials* 80 (2025): 104402.
155. J. Ren, H. Zhu, Y. Fang, et al., "Typical Cathode Materials for Lithium-Ion and Sodium-Ion Batteries: From Structural Design to Performance Optimization," *Carbon Neutralization* 2, no. 3 (2023): 339–377.
156. N. Wu, J. Shen, X. Zhou, et al., "Constructing Iron Vacancies in Thiospinel FeIn₂S₄ to Modulate Fe d-Band Center and Accelerate Sodiation Kinetics Enabling High-Rate and Durable Sodium Storage," *Advanced Energy Materials* 15, no. 19 (2025): 2405729.
157. Y. Liu, X. Rong, R. Bai, et al., "Identifying the Intrinsic Anti-Site Defect in Manganese-Rich Nasicon-Type Cathodes," *Nature Energy* 8 (2023): 1088–1096.
158. W. Zou, J. Li, R. Wang, et al., "Hydroxylamine Mediated Fenton-Like Interfacial Reaction Dynamics on Sea Urchin-Like Catalyst Derived From Spent LiFePO₄ Battery," *Journal of Hazardous Materials* 431 (2022): 128590.
159. J. M. F. Lucas, P. R. Prezas, S. Soreto Teixeira, et al., "Tuning the Magnetic and Electric Behavior of Lithium Ferrite Using an Eco-Friendly Pectin Sol-Gel Route," *Journal of Sol-Gel Science and Technology* 98 (2021): 580–592.
160. C. Wang, X. Kong, L. Wang, et al., "Mechanistically Engineered Heterojunction from Spent LFP for Efficient Oxygen Evolution Electrocatalysis," *Angewandte Chemie International Edition* 64, no. 43 (2025): e202516122.
161. X. Zhang, T. Liu, F. Zhao, N. Zhang, and Y. Wang, "In-Situ-Formed Cd and Ag₂S Decorated Cds Photocatalyst With Boosted Charge Carrier Spatial Separation for Enhancing Uv-Vis-Nir Photocatalytic Hydrogen Evolution," *Applied Catalysis, B: Environmental* 298 (2021): 120620.
162. Z. Yang, X. Xu, X. Liang, et al., "MIL-53(Fe)-Graphene Nanocomposites: Efficient Visible-Light Photocatalysts for the Selective Oxidation of Alcohols," *Applied Catalysis, B: Environmental* 198 (2016): 112–123.
163. Argonne National Laboratory (ANL). EverBatt: A Closed-Loop Battery Recycling Cost and Environmental Impacts Model, accessed, April 2019, <https://publications.anl.gov/anlpubs/2019/07/153050.pdf>.

164. J. Liu, Z. Liu, Z. Xiao, et al., "Iodine-Mediated Redox Strategy for Sustainable Lithium Extraction From Spent LiFePO_4 Cathodes," *Advanced Materials* 37, no. 26 (2025): 2503450.
165. G. Wang, T. Fearn, T. Wang, and K. L. Choy, "Machine-Learning Approach for Predicting the Discharging Capacities of Doped Lithium Nickel-Cobalt-Manganese Cathode Materials in Li-Ion Batteries," *ACS Central Science* 7, no. 9 (2021): 1551–1560.
166. M. Alyoubi, I. Ali, and A. M. Abdelkader, "Optimising the Regeneration Process of Spent Lithium-Cobalt Oxide Cathode Through Performance Analysis Model," *Journal of Energy Storage* 110 (2025): 115132.
167. J. Tang, H. Qu, C. Sun, et al., "A Universal Solution for Direct Regeneration of Spent Lithium Iron Phosphate," *Advanced Materials* 37, no. 14 (2025): 2420238.