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Challenges in the direct lithiation of spent LFP cathodes: the crucial role of reducing agents

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Recycling spent lithium-ion batteries (LIBs) has become essential for mitigating resource shortages and reducing the environmental impact. Especially with the wide use of $LiFePO_4$ cathodes, LIBs have become one of the most attractive batteries owing to their excellent electrochemical performance, long life cycle, safety and abundant availability of component elements. This article reviews the most advanced recycling technology for spent LFP cathodes: direct chemical regeneration. The literature review presented here focuses on the role of reducing agents. The direct recycling methods aim at repairing degraded LFP cathode materials and thereby reducing energy and chemical needs compared with traditional recycling methods, which are hindered by high energy consumption and secondary pollution. These innovations improve battery cycle life, lower energy consumption and production costs and support sustainable resource management. Direct regeneration technologies offer new opportunities to minimise resource waste and environmental pollution, promoting sustainable management of spent LIBs and advancing renewable energy technologies.

Broader context

This review addresses the pressing issues of resource depletion and environmental impact through the sustainable recycling of spent lithium-ion batteries (LIBs). Specifically, we focus on direct chemical regeneration of $LiFePO_4$ cathodes, a promising approach that reduces energy consumption, chemical usage, and waste production compared with traditional recycling methods. By exploring greener reducing agents, our work highlights the potential for more sustainable and environmentally friendly processes in the recycling industry. These innovations not only enhance the efficiency of battery recycling but also contribute to sustainable resource management, promoting a circular economy for renewable energy technologies.

1. Introduction

The rapid advancement in electric vehicle (EV) technology[1,2] has significantly increased interest in lithium iron phosphate ($LiFePO_4$)-based lithium-ion batteries (LIBs).[3,4] $LiFePO_4$ (LFP) with an olivine structure offers several remarkable properties, such as excellent thermal stability, long cycle life,[5–7] and inherent safety advantages, over other cathode materials such as NMC. In fact, olivine LFP ensures robust performance even under high temperature and pressure conditions, reducing the risk of thermal runaway and fire hazards, which are more common with other LIB chemistries.[8] Moreover, LFP is 32% cheaper than cobalt-based cathode materials.[9] These attributes make it an ideal candidate that meets the rigorous requirements of EV applications, in which reliability and safety are of paramount importance.[10] Its stability not only enhances the safety of EVs, but also contributes to the long-term durability of batteries, which is critical for the longevity of EVs and the sustainability of their large-scale adoption. Indeed, the production and deployment of LIBs featuring LFP-based cathodes rose from 27% in 2019 to 46% in 2022, with a projected growth of up to 64% by 2025.[10] Therefore, the importance of efficient recycling processes cannot be neglected. Spent batteries represent a valuable source of critical raw materials, including lithium, cobalt and nickel, which can be recovered and reused.[11] This concept of "urban mining" transforms end-of-life (EOL) batteries into a new source of chemical elements, offering significant environmental and economic benefits. By closing the loop through effective recycling, the dependency on virgin raw materials can be reduced, the environmental impact of mining activities can be mitigated, and a more sustainable supply chain for battery production can be built.[12,13] Table 1 summarises a non-comprehensive list of recycling actors and the recycling processes they use.[14]

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Table 1 List of companies and organizations in the battery recycling industry and the principal recycling processes they use[14]

Company	Process
SNAM (France)	Pyro, hydrometallurgy, direct recycling
Umicore (Belgium)	Pyro, hydrometallurgy
Sumimoto (Japan)	Pyro, hydrometallurgy
Accurec (Germany)	Pyrometallurgy
Dowa (Japan)	Pyro, hydrometallurgy
Brunp (China)	Pyro, hydrometallurgy
JX NMM (Japan)	Pyrometallurgy
MTB (France)	Pyrometallurgy
Retriev (Canada)	Hydrometallurgy
Recupyl (France)	Hydrometallurgy
SGE-HT (China)	Hydrometallurgy
Batrec (Switzerland)	Pyro, hydrometallurgy
OnTo technology (USA)	Direct recycling
Farasis energy (USA, Germany)	Direct recycling
LG energy (Korea)	Pyro, hydrometallurgy

The global market for LFP batteries is expanding rapidly, driven by rising EV adoption and renewable energy storage solutions.[15] In particular, the European Union (EU) has been actively promoting policies and initiatives to support the transition to clean energy and sustainable transportation. As a result, Europe is emerging as a significant player in the global battery supply chain, with substantial investments in battery manufacturing and recycling infrastructure.[16] To substantially reduce the carbon footprint of EV batteries, the EU regulatory framework aims to enforce specific recycling requirements by 2035, including mandatory minimum amounts of recycled material in new batteries: at least 20% for cobalt, 10% for lithium and 12% for nickel,[16] thus supporting both environmental and economic goals. Additionally, for efficient battery recycling, all the components should be considered, including cathode materials, anodes (*e.g.*, graphite),[17,18] electrolytes, and binders (*e.g.*, PVDF). Although reusing electrolytes is challenging due to degradation and solvent volatility, efforts are underway to improve electrolyte recycling from used batteries.[19]

Currently, pyrometallurgy and hydrometallurgy are the main approaches used to recycle spent LIBs.[20–23] Pyrometallurgy involves treating the batteries at very high temperatures, followed by separation steps that target the recovery of metals or alloys.[24,25] Hydrometallurgy includes a leaching step in an acidic solution to dissolve valuable metals present in spent LIBs and recover their salts or metallic hydroxides.[26–28] These two methods are widely applied in this industry.

Direct recycling has recently emerged as an alternative process to directly restore the active materials contained in spent batteries. It is a non-destructive material regeneration method enabling their reuse in new batteries[3,29] and is mainly applied at the laboratory scale today. More progress is expected in the coming years in order to develop greener and cost-effective industrial recycling processes. In fact, energy consumption and greenhouse gas (GHG) emissions are key parameters in the development of green and efficient recycling processes. On average, hydrometallurgical and pyrometallurgical processes consume 12.14 and 19.57 MJ kg_{cell}^{-1} of energy, respectively, primarily because of their multi-step recycling methodologies, high-temperature smelting and acid leaching with metal extraction/precipitation. In comparison, direct recycling requires only 3.16 MJ kg_{cell}^{-1}. The total GHG emissions are 1.96, 1.49, and 0.59 MJ kg_{cell}^{-1} for pyro-, hydro-, and direct recycling, respectively.[30] Even though these figures remain to be confirmed at higher technology readiness levels (TRL), direct recycling promises the lowest energy consumption and GHG emissions among the three recycling processes.[29]

In the case of LFP, recycling using standard industrial pyrometallurgy or hydrometallurgy methods is often cost-ineffective because the price of iron[31] is much lower than that of cobalt[32] or nickel[31] contained in NMC cathodes. Considering all the steps involved in these processes, as well as the required chemicals and high-temperature heating, the cost of a recycled LFP electrode can be higher than that of a new one. Hence, only direct recycling is considered as a viable process for recycling LFP from spent cathodes for reuse in new batteries.[33–36] Compared with pyro- and hydro-recycling methods, direct recycling provides significant benefits in terms of energy consumption, safety, cost, flexibility, and economic returns, thus garnering substantial attention from both academia and industry. In this context, researchers engaged in this field have developed several recycling methods for both spent batteries and electrode scraps.[37] This is evident from the exponential growth of the number of research articles on LFP recycling in the last few years (Fig. 1a).[38] The actual and projected growth in EVs[39,40] clearly demonstrates the concerted international focus on establishing a circular economy for battery materials.[15,41] Notably, for now, production scrap remains the primary source of battery waste as EOL battery volumes are not yet sufficient to sustain large-scale industrial recycling (Fig. 1b).

Despite the many advantages of direct recycling, this method remains more effective only at the laboratory scale, where high-purity cathode and anode material powders are hand-separated from the spent batteries. It is recognised, however, that a crucial aspect of industrial recycling is the production of black mass, a complex mixture of materials recovered after dismantling used batteries; it usually contains both electrode materials, carbon, and impurities from the Al and Cu current collectors. From this mixture, the direct recovery of pure electrode materials becomes extremely challenging due to the need for preliminary separation steps that allow the removal of all impurities before the regeneration steps.

To restore materials recovered from spent batteries, which may have degraded through different failure mechanisms discussed in the following section, various direct regeneration methods have been reported in the literature. These include solid-state sintering, electrochemical lithiation, and chemical lithiation in aqueous solutions by hydrothermal methods or in organic solvents by solvothermal methods.[10,29,40] In the specific case of LFP cathodes, direct recycling through chemi-

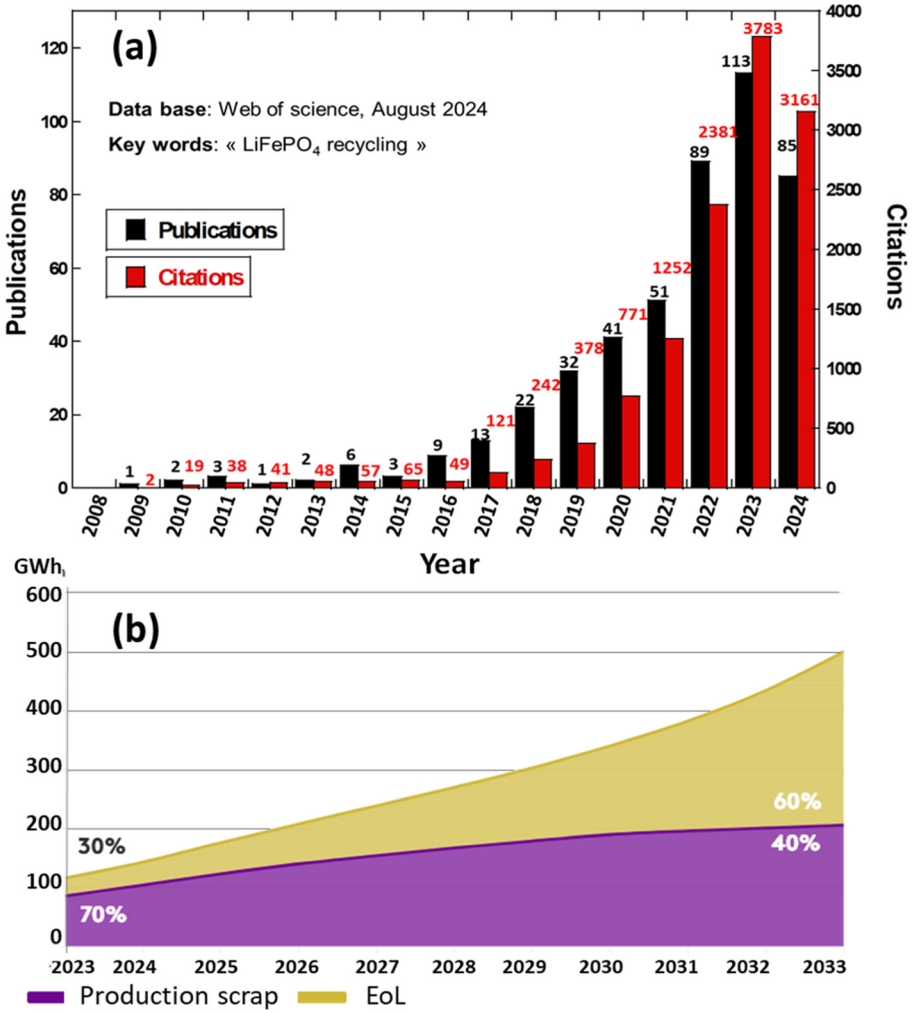

Fig. 1 Distribution of LFP recycling-related research articles from the Web of Science core collection by year, from 2008 to August 2024. The search term used was "$LiFePO_4$ recycling" (top). Scrap battery supply forecast (bottom).[42]

cal relithiation is a promising approach to addressing compositional and structural defects. This method allows the production of regenerated LFP that can be directly used in new battery assembly, eliminating the need to resynthesise new cathode materials from the recovered precursors.

This review does not aim to list the various regeneration methods available in the literature[10,29,43,44] but rather emphasises the importance of reducing agents. It briefly outlines the regeneration methods reported for LFP, highlighting the crucial role of reducing agents in the regeneration process. (Re)lithiation, in fact, is significantly affected by the type of reducing agent used as it promotes the reduction of Fe(III) in $FePO_4$ to Fe(II) in $LiFePO_4$ in the presence of a lithium source (Table 2). Thoroughly understanding these processes is vital for enhancing battery recycling techniques and developing more efficient and environment-friendly methods. To this end, the following sections examine various reducing agents and their roles in optimising the regeneration process of LFP batteries.

2. Why direct recycling through lithiation?

While conventional recycling methods are agnostic to the operational history of a battery, effective direct recycling necessitates a deep understanding of battery degradation mechanisms. Analogous to any repair process, identifying the root causes of failure is paramount to developing efficient and cost-effective regeneration strategies. Generally, battery failure can be caused by one or a combination of these phenomena: (i) current collector degradation, such as aluminum corrosion or copper dissolution; (ii) structural disorders, such as Li/Fe antisite defects in the spent LFP.[64] This is generally caused by the migration of Fe atoms of LFP from their M2 position to the Li site in the M1 position,[33] affecting Li^+ diffusion during charge/discharge cycling and thus leading to poor electrochemical performance and low reversible capacity;[56,65] (iii) particle cracking and carbon coating deterioration following multiple

Table 2 Representative relithiation methods reported for the LFP cathode material

Lithiation method	Reducing agent	Lithium source	Solvent	Conditions	Electrochemical performance	Ref.
Solid-state sintering	Ar/H_2	Li_2CO_3	—	650 °C/1 h	147.3 mAh g^{-1} at 0.2 C	45
	PVDF	Li_2CO_3	—	700 °C/3 h/Ar	151.55 mAh g^{-1} at 0.2 C	46
	Glucose	Li_2CO_3	—	300 °C/4 h 700 °C/10 h/Ar	146.89 mAh g^{-1} at 1 C	47
	Glucose	Li_2CO_3	—	750 °C/8 h/Ar	167.8 mAh g^{-1} at 0.1 C	48
	Sucrose	CH_3COOLi	—	800 °C/20 s/Ar	152 mAh g^{-1} at 0.1 C	49
Hydrothermal lithiation/annealing	Citric acid	LiOH Li_2CO_3	H_2O	80 °C/5 h 400–800 °C/1–7 h/N_2	159 mAh g^{-1} at 0.5 C	33
	Ascorbic acid	Li^+	H_2O	80 °C/2 h 600 °C/2 h/Ar	166.4 mAh g^{-1} at 0.1 C	50
	DL-Malic acid	LiOH	H_2O	100 °C/6 h 650 °C/6 h/Ar	138.4 mAh g^{-1} at 1 C	51
	Tartaric acid	LiOH	H_2O	200 °C/3 h 700 °C/2 h/Ar	145.92 mAh g^{-1} at 1 C	52
	Na_2SO_3	LiOH	H_2O	150 °C/24 h 600 °C/4 h/N_2	144.02 mAh g^{-1} at 0.1 C	53
	H_2O_2	LiOH	—	30 °C/1 h 400–800 °C/2–10 h/Ar	136.2 mAh g^{-1} at 1 C	54
Hydrothermal lithiation	Phytic acid	LiOH	H_2O	200 °C/5 h	133.6 mAh g^{-1} at 2 C	55
	Na_2SO_3	Li_2SO_4	H_2O	200 °C/6 h	145.1 mAh g^{-1} at 0.1 C	56
	$N_2H_4{\cdot}H_2O$	Li_2SO_4	H_2O	200 °C/3 h	141.9 mAh g^{-1} at 1 C	57
Chemical lithiation in organic solvents	Polycyclicaryl – lithium		Ethylene glycol dimethylether	Ambient T/10 min	160.1 mAh g^{-1} at 0.5 C	58
	- Ascorbic acid - Catechol - Hydroquinone - Oxalic acid - Glucose	$LiOAc{\cdot}2H_2O$:3EG		Ambient T/1–3 h	120 mAh g^{-1} at 0.5 C	59
	LiI		Acetonitrile	100 °C/1 h	150 mAh g^{-1} at 0.1 C	60
	LiI		Ethanol	Ambient T/48 h	168 mAh g^{-1} at 0.1 C	61
	LiI		—	Ambient T/10 min	168 mAh g^{-1} at 0.1 C	62
Molten salt regeneration	Sucrose	$LiNO_3$	—	300 °C/1–6 h/Ar 650 °C/6 h	145 mAh g^{-1} at 0.5 C	63

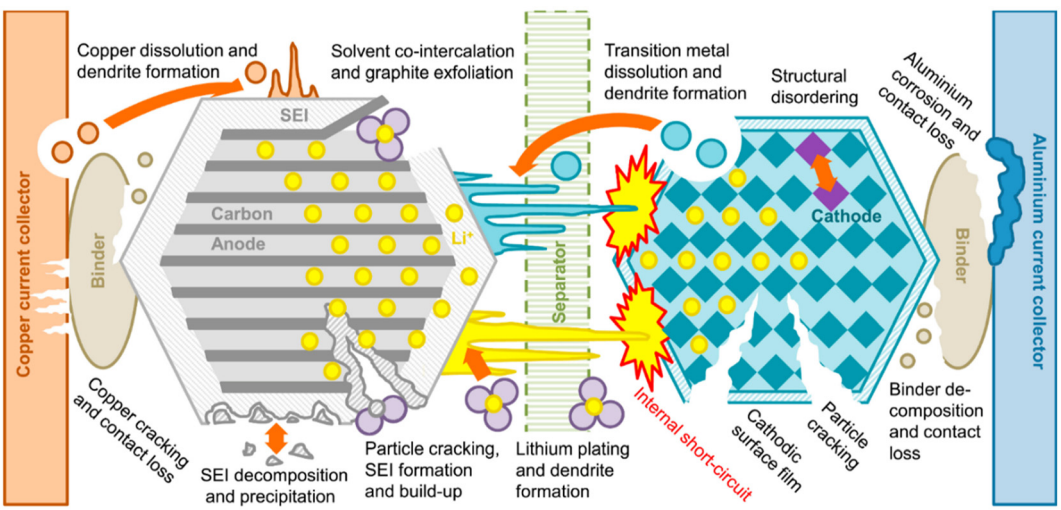

Fig. 2 Degradation mechanisms in LIBs (adapted from ref. 67).

charge–discharge cycles;[30,66] (iv) lithium inventory loss (Fig. 2).[67,68] In the case of LFP batteries, the major cause of EOL is the irreversible loss of lithium, primarily consumed for the formation and growth of the Solid Electrolyte Interphase (SEI) during charge/discharge cycling of the cell.[30,33,57,69] During the first few cycles, a certain amount of lithium ions from the cathode material is consumed along with the decomposition of the electrolyte, leading to the formation of a passivation layer on the surface of the anode (SEI) and the cathode (also named Cathode Electrolyte Interphase, CEI).[68]

These layers are relatively unstable and develop over time, which implies a continuous loss of lithium during cycling. SEI growth on the anode surface can increase interfacial overpotential, and when the SEI becomes unstable due to large volume fluctuations, it favours dendritic lithium plating. This promotes further lithium isolation and irreversible capacity loss. In the case of LFP, this is directly proportional to the amount of delithiated $FePO_4$ at the end of the discharge, which corresponds to the amount of lithium lost from the cathode.[70,71]

Therefore, this fraction of lithium lost cannot be recovered even at low discharge voltages (0 V). In other words, a fraction of Fe(III) cannot be reduced during electrochemical discharge. The aim of the lithiation process is to reduce the Fe(III) present in the FP phase to Fe(II) by compensating the missing lithium needed to form LFP. Therefore, it is of great importance to determine the exact amount of lithium to be supplemented during the regeneration process of the spent LFP in order to reduce the regeneration cost by supplying the appropriate amounts of reducing agent and lithium source.

3. Reducing agents for LFP relithiation

3.1. Reducing agent for high-temperature dry processes

Historically, solid-state sintering is the first reported lithiation process for spent LFP and is inspired by LFP synthesis methods. Generally, it involves heating a homogeneous mixture of spent LFP, a lithium source and a reducing agent at 600–900 °C under a controlled atmosphere.

The reducing agent generally consists of H_2 or carbonaceous materials (carbothermal method). H_2 has been used earlier to complete LFP synthesis in solution under a controlled atmosphere of N_2/H_2 (10% H_2) at 500 °C.[72] Although hydrogen is very useful for material synthesis, its significant drawback is that its extraction is a very complex process. The carbothermal process often involves readily available natural carbon or synthetic carbon. It can also be performed using organic compounds such as glucose or sucrose, which generate carbon *via* an *in situ* reaction. This carbon is not only useful for the carboreduction reaction but also generates a carbon coating on the LFP particles. Other carbon sources, such as CO/CO_2, can also be used.[73] Examples of reducing agents used in the LFP recycling field are discussed below; these include species that can directly regenerate LFP in the presence of a lithium source and those that work with recovered FP and Li_2CO_3 from spent cathodes as the lithium source in solution. Some works reported direct regeneration under pure nitrogen, with the binder playing the role of the carbon source.[46]

X. Li *et al.*[45] conducted a study on LFP cathode regeneration using spent LFP mixed with Li_2CO_3 at different temperatures from 600 to 800 °C under Ar/H_2 flow. The best results were obtained at 650 °C in this reductive atmosphere, and the regenerated cathode material mixture displayed excellent electrochemical performance even at 1 C. Fig. 3a shows that the material displayed the best discharge capacity and coulombic efficiency of 147.3 mAh g^{-1} at 0.2 C rate and 92.96%, respectively. Notably, J. Li *et al.*[46] reported a facile regeneration process in which Li_2CO_3 was first added to spent LFP, followed by ball milling for 6 h and subsequent heating at 700 °C for 3 h under an N_2 atmosphere. The reaction was conducted in the presence of C formed from the decomposition of PVDF according to the reaction: $[C_2H_2F_2]_n \rightarrow nHF_{(g)} + nC$. The regenerated LFP exhibited a discharge specific capacity of 151.55 mAh g^{-1} at a 0.2 C rate, qualifying this process as a facile and large-scale recycling method for LIBs.

B. Chen *et al.*[47] put forward a carboreduction process involving the addition of glucose as a source of carbon. Indeed, glucose and other carbohydrates can be decarbonated under an inert atmosphere starting from 400 °C (ref. 74) and then act as a reducing agent. At the same time, since it is well-known that LFP has poor electronic and ionic conductivity, the use of carbohydrates is also beneficial to improving its electrochemical performance by forming a carbon coating on the LFP particles. In the carbothermal reduction method highlighted in this study, Li_2CO_3 recovered from the spent LFP battery was used as the Li source (Fig. 3b); the mixture was heated to 300 °C for 4 h under argon and then annealed at 700 °C for 10 h. Sample LFP-G12 with a carbon content of 12 wt% in the LFP composite displayed better electrochemical properties, with an initial discharge capacity of 146.89 mAh g^{-1} at 1 C. Moreover, TEM and HRTEM analyses demonstrated that it contained a uniform carbon coating layer with a thickness of 2–3 nm. The use of glucose to provide C as a reducing agent at high temperatures was also endorsed by K. Liu *et al.*[48] who proposed the use of $H_4P_2O_7$ as the leaching acid for one-step selective recovery of metals from spent LFP batteries. In their work, the LFP was regenerated after recovering Li_2CO_3 and $FePO_4$ from the spent cathode using different leaching, precipitation and heat treatment steps (Fig. 3c). After mixing the recovered materials by ball milling for 3 h with 10 wt% glucose, the mixture was heated at 750 °C for 8 h under argon flow to regenerate spent LFP, which exhibited good electrochemical performance compared with the commercial cathodes, delivering 167.8 mAh g^{-1} initial capacity at 0.1 C and retained 91.31% after 100 cycles. In the same lines, another reducing agent from the same family was highlighted by S.–H. Zheng *et al.*[49] who proposed a strategy for rapid regeneration of spent LFP in just 20 s (Fig. 3d). This process was carried out by mixing lithium acetate as the lithium source and sucrose, which delivered C as the reducing agent; after 12 h of ball milling, the mixture was calcinated at 800 °C for 20 s under Ar. The results demonstrated that the structure and carbon layer of the spent LFP were well-restored after the regeneration process, which led to a significant improvement in its electrochemical activity and reversibility, achieving an initial capacity of 152 mAh g^{-1} at 0.1 C.

3.2. Reducing agents in aqueous solution

Aqueous solutions of carboxylic acids, such as citric and ascorbic acids, have been widely used to reduce Fe^{III} to Fe^{II}

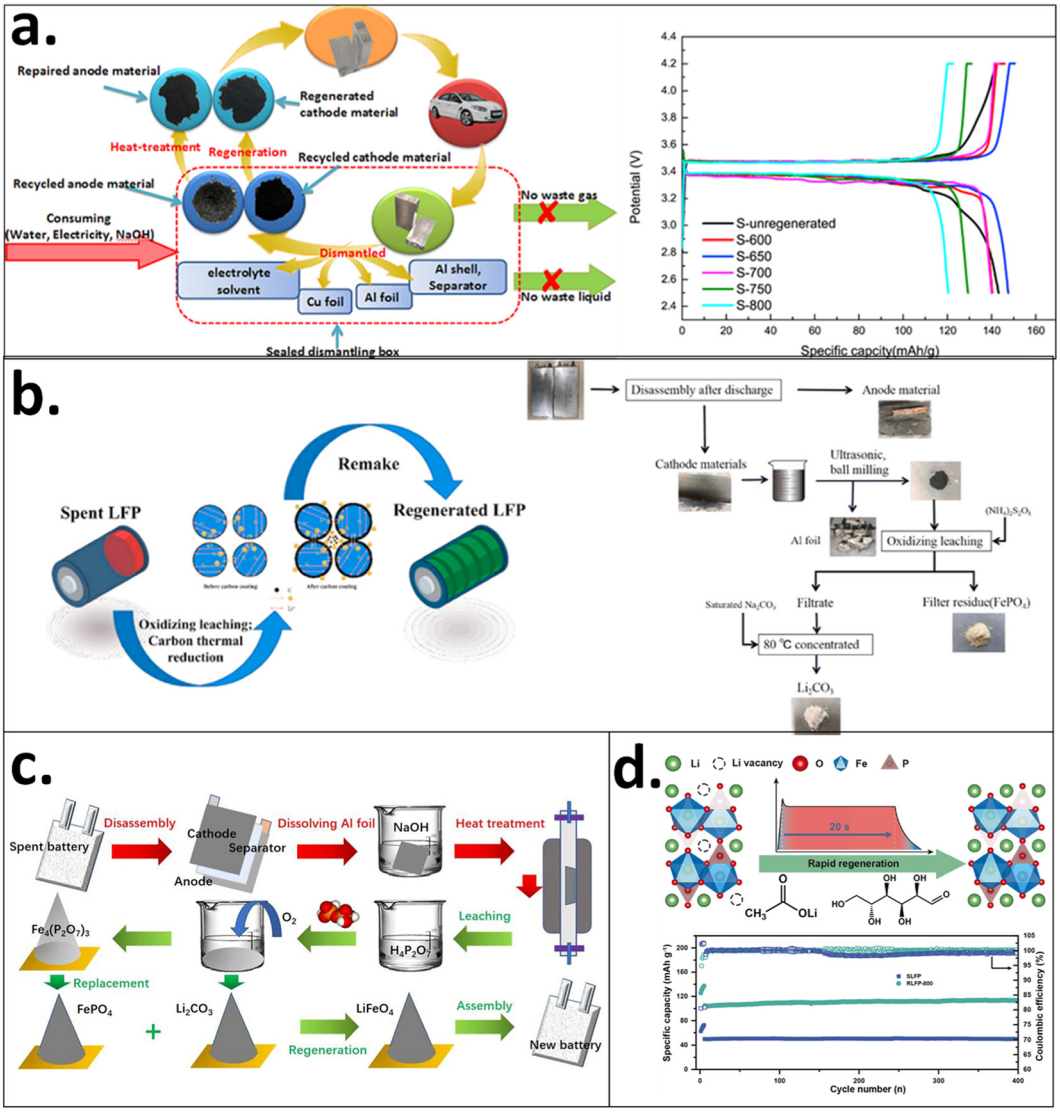

Fig. 3 Schematic representations of LFP regeneration *via* solid state sintering (a) using Li_2CO_3 and heating from 600 to 800 °C for 1 h under Ar/H_2 flow,[45] (b) by adding glucose and heating at 700 °C,[47] (c) by applying $H_4P_2O_7$ as the leaching acid and regenerating spent LFP by heating the recovered FP and Li_2CO_3 with glucose at 750 °C for 8 h (ref. 48) and (d) with an ultrafast regeneration strategy with heating only for 20 s at 800 °C using sucrose as the reducing agent.[49]

during the lithiation reaction by a hydrothermal process for direct LFP regeneration. Additionally, hydrazine is also widely used for LFP regeneration, similar to ascorbic acid or sugars during the synthesis of LFP in order to prevent the formation of ferric ions in aqueous media.[75] Examples of spent LFP regeneration in aqueous solutions with different reducing agents are illustrated below; this process typically includes heat treatments to restore the LFP structure.[75]

Among the various reducing agents, citric acid is discussed as a representative acid here. It was employed by P. Xu *et al.*[33] for spent LFP regeneration, as schematically shown in Fig. 4a. The process included a combination of aqueous solution relithiation of the spent LFP using 0.2 M LiOH and 0.08 M citric acid as the reducing agent; the lithiation reaction was performed at 80 °C for 5 h. Then, a post-annealing step was carried out by mixing Li_2CO_3 with the LFP powders and heating at 400–800 °C for 1–7 h under N_2. The obtained relithiated LFP exhibited good electrochemical performance, reaching an initial capacity of 159 mAh g^{-1}, while pristine LFP demonstrated a capacity of 161 mAh g^{-1} at 0.5 C. Furthermore, M. Fan *et al.*[50] proposed the restoration of degraded LFP *via* a hydrothermal process using lithium extracted from the spent anodes and ascorbic acid as the reducing agent, demonstrating the concept of treating waste with waste for the time. After 2 h of reaction at 80 °C, the obtained LFP was filtered and annealed at 600 °C for 2 h under an Ar atmosphere (Fig. 4d). The regenerated LFP delivered an improved discharge capacity of 166.4 mAh g^{-1} at a 0.1 C rate. The main advantage of this process is the recovery of lithium from spent anodes. J. Yang *et al.*[51] reported the use of DL-malic acid (a dicarboxylic acid)

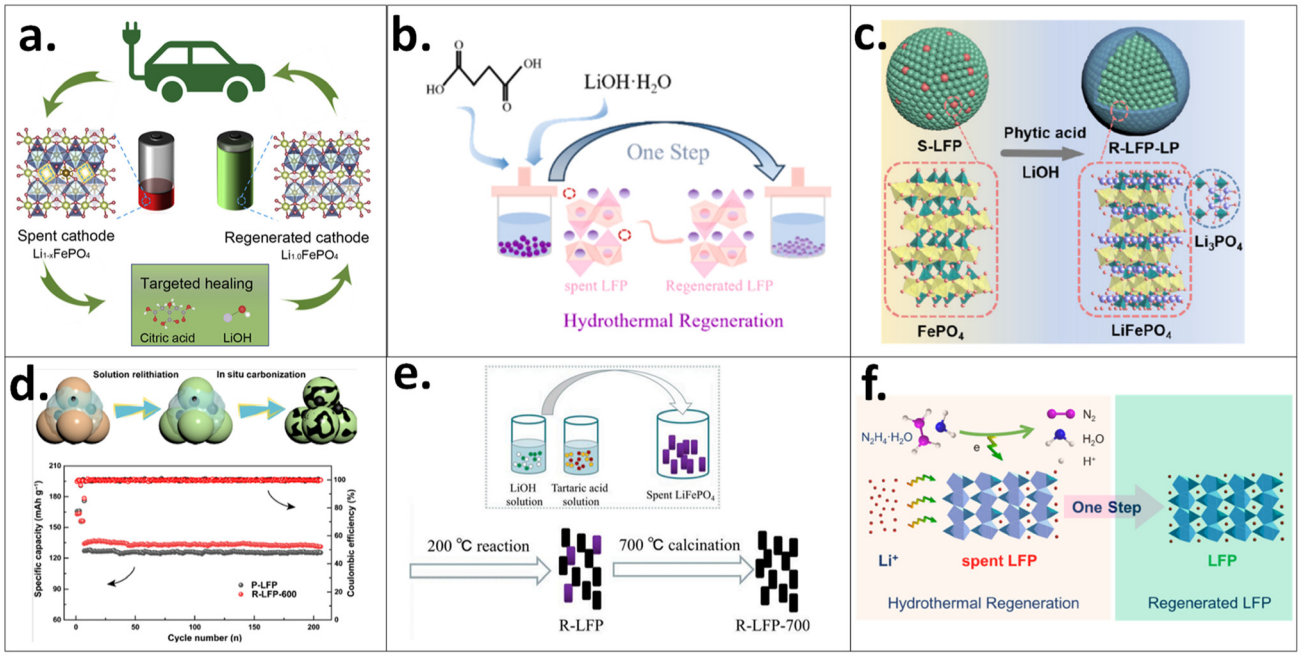

Fig. 4 Schematic representations of (a) LFP relithiation using citric acid,[33] (b) hydrothermal LFP regeneration process using malic acid,[51] (c) hydrothermal LFP lithiation using phytic acid,[55] (d) structural restoration of degraded LFP cathodes using residual Li from spent graphite anodes and ascorbic acid,[50] (e) the use of tartaric acid as a reducing agent for the hydrothermal lithiation of spent LFP,[52] and (f) hydrothermal LFP regeneration using $N_2H_4{\cdot}H_2O$ in an Li_2SO_4 solution.[57]

as a reducing agent for the direct regeneration of spent LFP by a hydrothermal process (Fig. 4b). Under optimal conditions, DL-malic acid was added to spent LFP with 1.2 mol L^{-1} LiOH. H_2O and heated at 100 °C for 6 h followed by 6 h of annealing at 650 °C. An initial discharge capacity of 138.4 mAh g^{-1} was obtained at 1 C, with good retention after 200 cycles.

An alternative organic acid has been highlighted recently by X. Zhu *et al.*[55] who reported a one-step hydrothermal process for spent LFP regeneration by using phytic acid as the reducing agent and LiOH as the lithium source at 200 °C for 5 h (Fig. 4c). They demonstrated that a nanolayer (9.6 nm) coating of Li_3PO_4 was formed on the LFP particles. Simultaneously, electrochemical measurements showed that the regenerated LFP possessed good electrochemical properties because of the Li_3PO_4 coating layer, resulting in 133.6 mAh g^{-1} at 2 C with a capacity retention of 94.86% after 300 cycles. Given this electrochemical performance, the 9.6 nm layer of Li_3PO_4 on the LFP particles does not seem to negatively impact the electronic conductivity of regenerated LFP.

The choice of reductant is all the more diverse; B. Chen *et al.*[52] developed a lithiation process using tartaric acid as the reductant along with LiOH under hydrothermal conditions of 200 °C for 3 h (Fig. 4e). Then, the regenerated material was annealed at 700 °C for 2 h under Ar. The obtained final material exhibited a high discharge capacity of 145.92 mAh g^{-1} at 1 C, with a capacity retention of 99.1% after 200 cycles.

Another type of reducing agent has also been suggested by X. Tang *et al.*[53] who used Na_2SO_3 to regenerate LFP by a hydrothermal reaction with LiOH at 150 °C for 24 h. The recovered material was mixed with Li_2CO_3 and annealed at 600 °C for 4 h to increase the crystallinity of lithiated LFP after hydrothermal treatment, leading to a specific capacity of 144.02 mAh g^{-1} at 0.1 C and a capacity retention of 92.36% after 100 cycles. Na_2SO_3 was also used as a reducing agent by Y. Yang *et al.*,[56] along with Li_2SO_4 as the lithium source, to restore spent LFP by a hydrothermal process performed for 6 h at 200 °C without an additional annealing step.

Q. Jing *et al.*[57] proposed a hydrothermal regeneration process by using $N_2H_4{\cdot}H_2O$ as a reducing agent (Fig. 4f) in Li_2SO_4 solution which was used as the lithium source. The obtained mixture containing the spent LFP was placed in a blast oven (inside an autoclave) at 200 °C for 3h. The regenerated LFP displayed good discharge capacity of 141.9 mAh g^{-1} at 1 C with a good capacity retention reaching 98.6% after 200 cycles. The advantage of this process is that it doesn't require a supplementary heating step which is less energy-consuming.

Another common reducing agent found in the literature is H_2O_2, which was put forward by Y. Xu *et al.*[54] for the direct recovery of degraded LFP *via* a mild chemical relithiation strategy; LiOH was added to spent LFP along with H_2O_2 as the reducing agent in an aqueous solution and reacted for 1 h at 30 °C in the air. After chemical relithiation, the obtained regenerated LFP was mixed with Li_2CO_3 and annealed at different annealing temperatures from 400 to 800 °C and for different calcination durations from 2 to 10 h under argon in order to obtain regenerated LFP with high crystallinity. The obtained RA-LFP (heated at 700 °C) exhibited a capacity of 136.2 mAh g^{-1} at 1 C and a capacity retention of 84.9% after 1000 cycles at 5 C.

3.3. Reducing agents used in organic solvents

Apart from hydrothermal lithiation, various reducing agents are used for the regeneration of EOL LFP cathodes through chemical relithiation in organic solvents. This process is carried out at atmospheric pressure, which offers a significant safety advantage.[10]

C. Wu *et al.*[58] proposed a direct chemical regeneration process in which polycyclicaryl-lithium compounds served as both reducing agent and lithium source, and ethylene glycol dimethylether was employed as the solvent (Fig. 5a). The chemical relithiation of spent LFP was achieved within 10 minutes under ambient temperature and pressure, and the obtained material exhibited a discharge capacity of up to 160.1 mAh g^{-1} at 0.5 C. Despite the potential advantages of this technique, the volatile organic solvents and the demanding synthesis of polycyclic aryl-lithium compounds used in this process require safety precautions to be followed and increase the cost of the entire process, especially for industrial applications.

T. Yingnakorn *et al.*[59] presented closed-loop recycling of spent LFP *via* direct lithiation under ambient temperature and pressure, using a eutectic system containing lithium acetate and ethylene glycol ($LiOAc{\cdot}2H_2O$:3EG) both as the solvent and the lithium source. Different organic reducing agents, including ascorbic acid, catechol, hydroquinone, oxalic acid, and glucose, were dissolved in the prepared solvent and tested. They proposed two regeneration routes, with and without the leaching of lithium ions in water using iron(III) chloride ($FeCl_3$) as the oxidising agent, followed by the same lithiation process described above. The solution containing spent LFP was stirred for 1 to 3 hours at 25 °C and then treated at 450 °C for 1 h under argon to eliminate the remaining polymer binder. The recovered LFP was then reformulated again for electrochemical testing, which revealed that the cathode exhibited a discharge capacity of 120 mAh g^{-1} at 0.5 C *vs.* Li^+/Li.

In fact, one of the most common reducing agents used for direct chemical lithiation of LFP in organic solvents is lithium iodide (LiI), which was first used in acetonitrile.[76,77] For instance, after dismantling the batteries, M. Ganter *et al.*[60] recovered the spent LFP by scraping it off the current collector and grinding it. The obtained LFP black mass was relithiated in a 1 M acetonitrile solution of lithium iodide, acting as both the reducing agent and the lithium source, stirred for 20 h in air. After washing with acetonitrile to remove excess LiI, the obtained LFP was dried at 100 °C for 1 h. The electrochemical analysis demonstrated total lithiation of LFP, which exhibited a good capacity of 150 mAh g^{-1} at 0.1 C. Based on this process, which requires the use of an organic solvent, especially for large-scale application, our team proposed a wide choice of solvents (acetonitrile, ethanol, cyclohexane, methanol, DMSO and propane-1,2-ol) that can be applied to chemically lithiate spent LFP using LiI at room temperature under air conditions.[61] The LFP regenerated in ethanol directly by placing

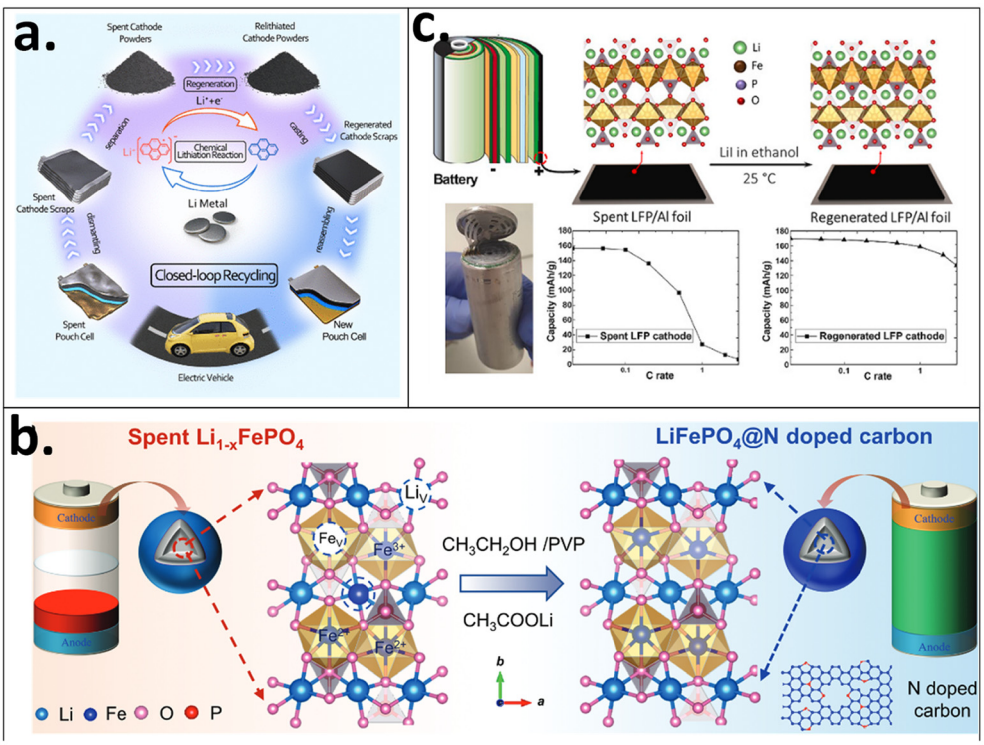

Fig. 5 Schematic representations of (a) direct chemical regeneration of LFP using polycyclic aryl-lithium compounds in ethylene glycol dimethyl ether,[58] (b) regeneration process using lithium acetate in ethanol,[64] and (c) direct LFP regeneration at room temperature in different organic solvents using lithium iodide.[61]

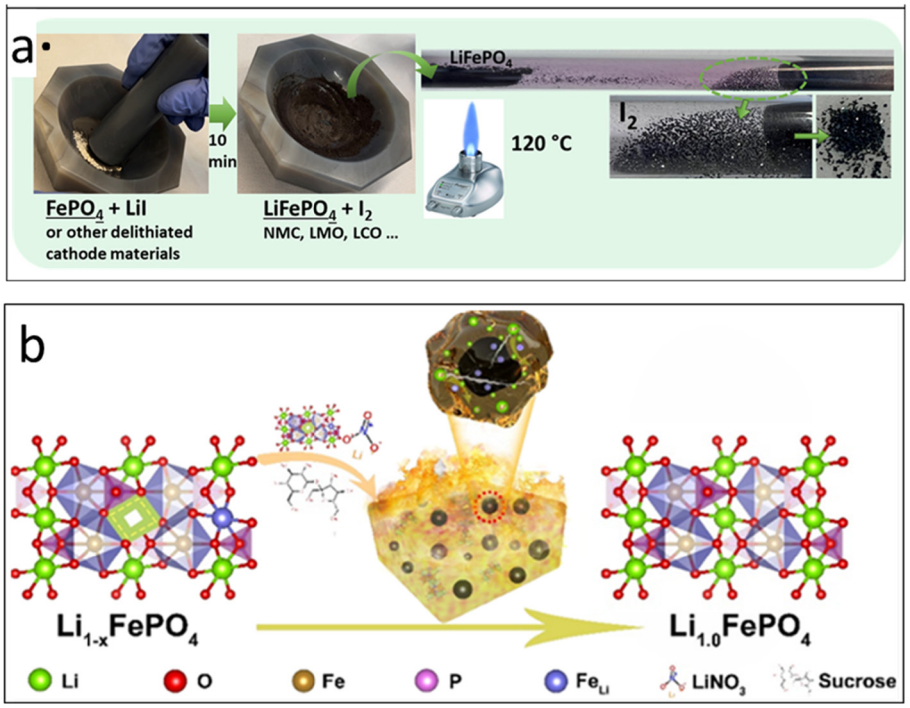

Fig. 6 (a) Solvent-free direct lithiation of spent LFP and other cathode materials at room temperature with very rapid reaction kinetics[62] and (b) direct LFP regeneration *via* a low-temperature molten salt process coupled with a reductive environment.[63]

the material cast onto its aluminum current collector demonstrated excellent electrochemical performance, proving advantageous since it is one of the greenest and cheapest solvents and avoids additional heat treatment. Moreover, the LFP regenerated in ethanol exhibited a full reversible capacity of 168 mAh g^{-1} at 0.1 C and good performance even at a high current density (Fig. 5c). This process preserved the electrode formulation and carbon coating of the LFP particles, and more importantly, the LFP cathode was regenerated at room temperature.

Dan Yang *et al.*[78] reported another strategy that involved soaking the spent $LiFePO_4$ electrodes directly in the LiTBH solution (Lithium triethyl borohydride/tetrahydrofuran Li $(C_2H_5)_3BH$/THF; LiTBH) for 6 minutes at room temperature. This process rejuvenated the crystal structure and electrochemical activity of the spent LFP electrodes.

3.4. Solvent-free methods

The works highlighted in the previous section show that lithiation using LiI works successfully in various solvents. This raises the question of the usefulness of a solvent for the lithiation of FP using LiI. For this reason, recently, our team reported a new solvent-free process (FSF) at ambient air and temperature conditions with fast reaction kinetics (Fig. 6a).[62] The spent LFP cathode powder was regenerated without separating the PVDF polymer binder and conductive carbon additive using LiI at a molar ratio of LiI/FP = 1 for an FP content of 30% in spent LFP. The mixture was hand-mixed for 10 minutes, and the regenerated LFP exhibited a reversible capacity of 168 mAh g^{-1}. This method holds the potential for seamless extension to other LIB cathode materials, including LMFP, LMO, LCO and NMC. Additionally, it offers the possible recovery of I_2 crystals by sublimation, such that it can be used in other applications.

An alternative method, which does not require an organic solvent or aqueous solution, is the molten salt method. It can also be used for the direct regeneration of LFP by employing eutectic molten salts in the reaction medium. A nitrite molten salt system can be applied to recycle spent cathode materials such as NMC,[79] but in the case of LFP, the oxidation of Fe^{2+} to Fe^{3+} by nitrite is more favourable. In order to tackle this issue, X. Liu *et al.*[63] proposed a direct regeneration process for spent LFP by using lithium nitrite as both the molten salt medium and the lithium source; with sucrose as the carbon source to provide a reductive environment,[80] the reaction was carried out at 300 °C, followed by annealing at 650 °C (Fig. 6b). Their findings demonstrated that the structure of LFP particles was recovered, and the regenerated material delivered an improved specific capacity of 145 mAh g^{-1} at 0.5 C. The regeneration strategy presented by X. Lv *et al.*[81] involves utilising photocatalysis to correct the anti-site defects by reducing the energy barrier for Fe migration (Fe^{3+} ions are reduced to Fe^{2+}, while Li^+ ions are incorporated into the LFP lattice, restoring the capacity of the material). The process is initiated by photocatalytic activation under light irradiation at ambient conditions, offering a sustainable and efficient means to recycle spent LFP cathodes.

4. Challenges and future solutions

Efficient direct recycling of spent LIBs involves several steps to properly prepare the batteries for the recycling process and, especially, recover the cathode and anode materials with high purity avoiding impurities. Starting from discharging the cells, which is primarily done for safety reasons but also to fully reduce the FP, in the case of LFP. This allows for the accurate determination of the percentage of missing lithium in the cathode. The current challenge is in finding the optimal discharge potential for the spent batteries before opening them. Fully discharging them to 0 V or short-circuiting can lead to copper oxidation, resulting in impurities in the recovered materials and poor cycling properties. Besides these serious challenges at the industrial level, certain other parameters, such as the choice of the reducing agent, are also crucial determinants of the direct recycling method, as mentioned above.

- **Critical perspectives on various direct LFP lithiation pathways**: In the case of LFP, which is the focus of this review, conventional methods cannot be applied for environmentally sustainable recycling of the active material, as mentioned previously, due to the relatively modest cost of pristine LFP. Direct recycling is more suitable for this purpose. In addition, as mentioned in the introduction, the challenge in direct recycling of spent LFP cathodes lies in the development of lithiation processes that are simple, cost-effective, and environmentally friendly. Importantly, the recycled LFP should not be costlier than the pristine materials to encourage recycling. With this in mind, several research teams have developed direct lithiation processes using various approaches (Table 2), such as solid-state sintering, chemical lithiation by hydrothermal method and chemical lithiation in organic solvents by solvothermal method, besides highlighting their advantages and limitations, particularly for large-scale industrial applications (Fig. 7).

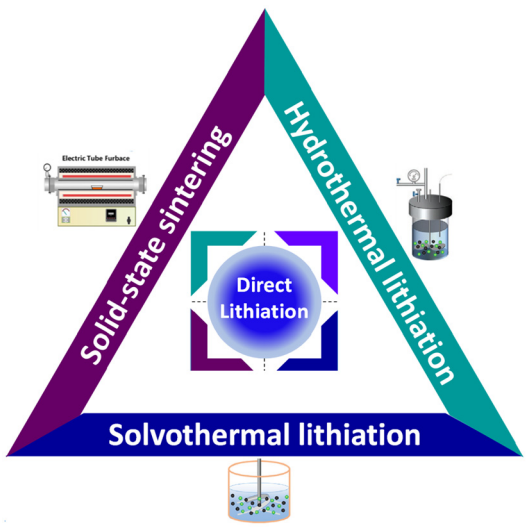

Fig. 7 Schematic representation of direct lithiation methods for spent LFP.

Solid-state sintering is a well-established method for material synthesis and has been used for decades. It has been widely applied in the production of ceramics, metals and composite materials, owing to its ability to produce high-density and uniform material structures. In the lithiation context, solid-state sintering is used not only due to its simplicity and scalability but, particularly, to enhance the electrochemical properties of the materials; moreover, it is potentially robust against impurities since the volatile components evaporate at high temperatures.[82–85] For LFP, the conditions used in solid-state sintering are similar to those in its synthesis processes. Although the sintering process takes less time than synthesis and involves a single calcination step, it still consumes a significant amount of energy.[29] The most promising solid-state method without solvent usage is the fast solvent-free (FSF) lithiation process, which has been developed using LiI at ambient temperature. The challenge here is how to manage the iodine formed on a large scale, which can be recovered through sublimation, but special care must be taken while handling I_2 vapors for safety. Alternatively, iodine can be captured in ethanol as I_3^- (which is used in Betadine).

Hydrothermal lithiation, which is largely reported for LFP cathode regeneration, requires a lithium source and a reducing agent to regenerate the spent LFP in aqueous solutions.[86] Its advantages include operational simplicity, minimum waste production and scalability. Above all, the use of water as a solvent is very beneficial for the direct regeneration of spent cathode materials. It requires a close system with an autoclave container that is heated in the temperature range of 80 °C to 200 °C; this generates autogenous pressure that can reach 15–20 bar in some cases, thus posing limitations in terms of security in addition to the need for autoclaves that require large investment at the large scale. Furthermore, this regeneration method generally requires a second annealing step at a high temperature to relax the structure and avoid structural defects related to the occupation of the M1 and M2 sites in the structure, which can be a real drawback in terms of energy consumption.

Finally, unlike hydrothermal processes, **solvothermal lithiation** can be conducted at atmospheric pressure and low temperatures. The main features of this method are as follows: (i) it allows regeneration of the entire cathode while preserving the conductive carbon and polymer binder along with the active material; (ii) it operates at (low) ambient temperatures and thus facilitates the insertion reaction without any structural effects, such as Fe and Li mixing. (iii) In addition, it is tolerant to a large choice of organic solvents, which increases its feasibility. Indeed, the need for an organic solvent impacts the price and the toxicity, which are hence considered the major disadvantages of this method. Moreover, the most promising solvent may be ethanol, even if it features slower reaction kinetics (48 h).

Based on the different lithiation methods discussed above and the conditions related to the use of a given reducing agent (Table 2), it appears that the key to these processes is the nature of the reducing agent used. In the future, it will be chal-

lenging to explore other reducing agents that do not require organic solvents and can operate at low temperatures. Alternatively, developing water-based methods using green reducing agents that are environmentally friendly and cost-effective would be beneficial. These methods should ideally operate at temperatures close to ambient conditions to prevent Fe/Li migration and avoid high-temperature heating steps.

- **Life cycle assessment**: LCA is crucial for the recycling processes as it provides a comprehensive evaluation of the environmental impacts associated with each stage. By systematically quantifying inputs, such as energy and raw materials, and output characteristics such as emissions and waste are evaluated. LCA employs a standardised methodology (ISO 14040-44) to ensure consistency and accuracy. This thorough analysis guides the development of direct recycling processes by highlighting areas where environmental benefits can be maximised and negative impacts can be minimised.[87] Moreover, LCA is crucial for these processes, especially when scaling them up for industrial applications, which can introduce new challenges and potential environmental impacts that are not straightforward at smaller scales. LCA ensures that large-scale operations remain sustainable and environmentally friendly. However, LCAs conducted at the laboratory scale are less relevant compared with those performed at the pilot or industrial scale. Ultimately, the application of LCA to recycling methods fosters sustainable practices in the management of spent LIBs, ensuring that the recycling methods are not only efficient but also pose minimal environmental harm. This is essential for the advancement of a circular economy and the reduction of the overall environmental footprint of battery usage.[88–90]

5. Summary and perspective

The direct recycling of LFP batteries is a promising approach to reduce environmental impact and resource consumption. This recycling process often involves the selection of an appropriate reducing agent to restore the electrochemical performance of the cathode. Various reducing agents, such as hydrogen, ascorbic acid, and carbohydrates, have been explored, each offering distinct advantages and challenges in terms of efficiency, cost, and environmental footprint. The ultimate goal in the field of LFP recycling is to develop a regeneration process that is economically competitive with pristine LFP production. Therefore, it is essential to streamline the recycling procedure, minimise handling steps and optimise the restoration of cathode material properties. Achieving this challenging goal would significantly enhance the sustainability of LFP battery recycling, making battery usage more economically viable and environmentally friendly. Ultimately, continued research and development of innovative recycling techniques that focus on the synergistic effects of reducing agents and lithium sources are critical to realising this vision. This approach has the potential to contribute significantly to the circular economy of battery materials and support the growing demand for energy storage solutions, while minimising waste and resource depletion.

In the literature, different regeneration processes have been proposed using various lithiation methods, such as solution-based and solid-state approaches, which are demonstrated using small quantities (a few grams) at the laboratory scale. These methods have mostly resulted in high-purity regenerated cathodes obtained from properly separated and recovered spent cathodes. The challenge now is to test these processes at the pilot scale, particularly with industrial black mass.

Notably, high-quality black mass can be obtained only through manual disassembly and separation, which are labor-intensive and require careful handling to avoid risks of explosion or leakage of toxic substances. Meanwhile, industrial methods use machines to crush and separate battery components. This process is fast and can handle large quantities of batteries, which is preferred for large-scale recycling that prioritises efficiency and processing capacity. However, the black mass obtained may contain more impurities due to less selective separation. Doubts about the relevance of direct recycling of LIBs are now less justified, since the poor quality of current industrial black mass may be overcome. Indeed, huge efforts[91,92] have been devoted to developing new and advanced automated processes to produce high-quality industrial black mass based on novel robotic[93] and artifical intelligence (AI) technologies (Fig. 8). For example, EU has funded many research programs that aim to develop such new methods (RECIRCULATE, Reuse,[94] Griner, and Eurecat). The importance of AI and machine learning in the recycling of batteries, particularly LFP batteries, is increasingly recognised by researchers today. These technologies are transforming the recycling process by enhancing efficiency and safety while reducing costs. Advanced AI systems optimise battery sorting and disassembly, predict necessary maintenance, and improve methods for the recovery of valuable metals, thus minimising environmental impact and reducing the need for mining.[95,96] These efforts are expected to lead to fully automated spent

Fig. 8 Integrating AI with leading technology pathways (from cicenergigune.com).

battery treatments. Finally, an optimised and well-chosen approach for each type of battery and black mass can maximise the economic and environmental benefits of direct recycling of LIBs.[97]

Conflicts of interest

There are no conflicts to declare.

Data availability

No primary research results, software or code has been included and no new data were generated or analysed as part of this review.

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References

1 R. Bird, Z. J. Baum, X. Yu and J. Ma, The Regulatory Environment for Lithium-Ion Battery Recycling, *ACS Energy Lett.*, 2022, **7**, 736–740, DOI: **10.1021/acsenergylett.1c02724**.
2 X. Zhang, L. Li, E. Fan, Q. Xue, Y. Bian, F. Wu and R. Chen, Toward sustainable and systematic recycling of spent rechargeable batteries, *Chem. Soc. Rev.*, 2018, **47**, 7239–7302, DOI: **10.1039/c8cs00297e**.
3 B. Zhang, Y. Xu, D. S. Silvester, C. E. Banks, W. Deng, G. Zou, H. Hou and X. Ji, Direct regeneration of cathode materials in spent lithium-ion batteries toward closed-loop recycling and sustainability, *J. Power Sources*, 2024, **589**, 233728, DOI: **10.1016/j.jpowsour.2023.233728**.
4 J. Zhang, J. Zou, D. He, W. Hu, D. Peng, Y. Li, Z. Zhao, S. Wang, P. Li, S. Su, K. Ma and X. Wang, Molten salt infiltration-oxidation synergistic controlled lithium extraction from spent lithium iron phosphate batteries: an efficient, acid free, and closed-loop strategy, *Green Chem.*, 2023, **25**, 6057–6066, DOI: **10.1039/d3gc00116d**.
5 A. K. Padhi, K. S. Nanjundaswamy and J. B. Goodenough, Phospho–olivines as Positive–Electrode Materials for Rechargeable Lithium Batteries, *J. Electrochem. Soc.*, 1997, **144**, 1188–1194, DOI: **10.1149/1.1837571**.
6 K. Zaghib, A. Mauger, J. B. Goodenough, F. Gendron and C. M. Julien, Electronic, Optical, and Magnetic Properties of $LiFePO_4$: Small Magnetic Polaron Effects, *Chem. Mater.*, 2007, **19**, 3740–3747, DOI: **10.1021/cm0710296**.
7 M. S. Whittingham, Lithium batteries and cathode materials, *Chem. Rev.*, 2004, **104**, 4271–4301, DOI: **10.1021/cr020731c**.
8 A. W. Golubkov, D. Fuchs, J. Wagner, H. Wiltsche, C. Stangl, G. Fauler, G. Voitic, A. Thaler and V. Hacker, Thermal-runaway experiments on consumer Li-ion batteries with metal-oxide and olivin-type cathodes, *RSC Adv.*, 2014, **4**, 3633–3642, DOI: **10.1039/c3ra45748f**.
9 O. Catsaros, *Lithium-Ion Battery Pack Prices Hit Record Low of $139/kWh|BloombergNEF, BloombergNEF*, 2023, pp. 1–4.
10 Y. Ding, J. Fu, S. Zhang, X. He, B. Zhao, J. Ren, J. Zhong and Z. Liu, Advances in recycling $LiFePO_4$ from spent lithium batteries: A critical review, *Sep. Purif. Technol.*, 2024, **338**, 126551, DOI: **10.1016/j.seppur.2024.126551**.
11 Y. Zhao, X. Yuan, L. Jiang, J. Wen, H. Wang, R. Guan, J. Zhang and G. Zeng, Regeneration and reutilization of cathode materials from spent lithium-ion batteries, *Chem. Eng. J.*, 2020, **383**, 123089, DOI: **10.1016/j.cej.2019.123089**.
12 J. Neumann, M. Petranikova, M. Meeus, J. D. Gamarra, R. Younesi, M. Winter and S. Nowak, Recycling of Lithium–Ion Batteries—Current State of the Art, Circular Economy, and Next Generation Recycling, *Adv. Energy Mater.*, 2022, **12**, 2102917, DOI: **10.1002/aenm.202102917**.
13 M. Armand and J. M. Tarascon, Building better batteries, *Nature*, 2008, **451**, 652–657, DOI: **10.1038/451652a**.
14 F. Larouche, F. Tedjar, K. Amouzegar, G. Houlachi, P. Bouchard, G. P. Demopoulos and K. Zaghib, Progress and status of hydrometallurgical and direct recycling of Li-Ion batteries and beyond, *Materials*, 2020, **13**, 801, DOI: **10.3390/ma13030801**.
15 X. Chen, W. Shen, T. T. Vo, Z. Cao and A. Kapoor, An overview of lithium-ion batteries for electric vehicles, in 10th Int. Power Energy Conf. IPEC 2012, IEEE, 2012, pp. 230–235, DOI: **10.1109/ASSCC.2012.6523269**.
16 Y. Jung *et al.*, EU Battery Regulation to Enter Into Force in 2024, Kim & Chang, **https://www.kimchang.com/en/insights/detail.kc?sch_section=4&idx=27042** (accessed April 16, 2025).
17 S. Natarajan and V. Aravindan, An Urgent Call to Spent LIB Recycling: Whys and Wherefores for Graphite Recovery, *Adv. Energy Mater.*, 2020, **10**, 2002238, DOI: **10.1002/aenm.202002238**.
18 Y. Gao, C. Wang, J. Zhang, Q. Jing, B. Ma, Y. Chen and W. Zhang, Graphite Recycling from the Spent Lithium-Ion Batteries by Sulfuric Acid Curing-Leaching Combined with High-Temperature Calcination, *ACS Sustainable Chem. Eng.*, 2020, **8**, 9447–9455, DOI: **10.1021/acssuschemeng.0c02321**.
19 R. Zhang, X. Shi, O. C. Esan and L. An, Organic Electrolytes Recycling From Spent Lithium-Ion Batteries, *Global Challenges*, 2022, **6**, 1–10, DOI: **10.1002/gch2.202200050**.
20 Z. Dobó, T. Dinh and T. Kulcsár, A review on recycling of spent lithium-ion batteries, *Energy Rep.*, 2023, **9**, 6362–6395, DOI: **10.1016/j.egyr.2023.05.264**.
21 M. Chen, X. Ma, B. Chen, R. Arsenault, P. Karlson, N. Simon and Y. Wang, Recycling End-of-Life Electric Vehicle Lithium-Ion Batteries, *Joule*, 2019, **3**, 2622–2646, DOI: **10.1016/j.joule.2019.09.014**.
22 Z. J. Baum, R. E. Bird, X. Yu and J. Ma, Lithium-Ion Battery Recycling–Overview of Techniques and Trends, *ACS Energy Lett.*, 2022, **7**, 712–719, DOI: **10.1021/acsenergylett.1c02602**.

23 M. Wang, K. Liu, S. Dutta, D. S. Alessi, J. Rinklebe, Y. S. Ok and D. C. W. Tsang, Recycling of lithium iron phosphate batteries: Status, technologies, challenges, and prospects, *Renewable Sustainable Energy Rev.*, 2022, **163**, 112515, DOI: **10.1016/j.rser.2022.112515**.

24 T. Georgi-Maschler, B. Friedrich, R. Weyhe, H. Heegn and M. Rutz, Development of a recycling process for Li-ion batteries, *J. Power Sources*, 2012, **207**, 173–182, DOI: **10.1016/j.jpowsour.2012.01.152**.

25 A. Holzer, S. Windisch-Kern, C. Ponak and H. Raupenstrauch, A novel pyrometallurgical recycling process for lithium-ion batteries and its application to the recycling of LCO and LFP, *Metals*, 2021, **11**, 1–22, DOI: **10.3390/met11010149**.

26 A. Chagnes and B. Pospiech, A brief review on hydrometallurgical technologies for recycling spent lithium-ion batteries, *J. Chem. Technol. Biotechnol.*, 2013, **88**, 1191–1199, DOI: **10.1002/jctb.4053**.

27 H. Mahandra and A. Ghahreman, A sustainable process for selective recovery of lithium as lithium phosphate from spent $LiFePO_4$ batteries, *Resour., Conserv. Recycl.*, 2021, **175**, 105883, DOI: **10.1016/j.resconrec.2021.105883**.

28 L. Zhang, T. Teng, L. Xiao, L. Shen, J. Ran, J. Zheng, Y. Zhu and H. Chen, Recovery of $LiFePO_4$ from used lithium-ion batteries by sodium-bisulphate-assisted roasting, *J. Cleaner Prod.*, 2022, **379**, 134748, DOI: **10.1016/j.jclepro.2022.134748**.

29 X. Qiu, C. Wang, L. Xie, L. Zhu, X. Cao and X. Ji, Challenges and perspectives towards direct regeneration of spent $LiFePO_4$ cathode, *J. Power Sources*, 2024, **602**, 234365, DOI: **10.1016/j.jpowsour.2024.234365**.

30 G. Ji, J. Wang, Z. Liang, K. Jia, J. Ma, Z. Zhuang, G. Zhou and H. M. Cheng, Direct regeneration of degraded lithium-ion battery cathodes with a multifunctional organic lithium salt, *Nat. Commun.*, 2023, **14**, 584, DOI: **10.1038/s41467-023-36197-6**.

31 National Minerals Information Center, Mineral Commodity Summaries 2024, DOI: **10.3133/mcs2024**.

32 Cobalt Institute, *Cobalt Market Report 2022*, 2023.

33 P. Xu, Q. Dai, H. Gao, H. Liu, M. Zhang, M. Li, Y. Chen, K. An, Y. S. Meng, P. Liu, Y. Li, J. S. Spangenberger, L. Gaines, J. Lu and Z. Chen, Efficient Direct Recycling of Lithium-Ion Battery Cathodes by Targeted Healing, *Joule*, 2020, **4**, 2609–2626, DOI: **10.1016/j.joule.2020.10.008**.

34 V. Gupta, M. Appleberry, W. Li and Z. Chen, Direct recycling industrialization of Li-ion batteries: The pre-processing barricade, *Next Energy*, 2024, **2**, 100091, DOI: **10.1016/j.nxener.2023.100091**.

35 G. Hu, Y. Gong, Z. Peng, K. Du, M. Huang, J. Wu, D. Guan, J. Zeng, B. Zhang and Y. Cao, Direct Recycling Strategy for Spent Lithium Iron Phosphate Powder: an Efficient and Wastewater-Free Process, *ACS Sustainable Chem. Eng.*, 2022, **10**, 11606–11616, DOI: **10.1021/acssuschemeng.2c03520**.

36 C. Qi, S. Wang, X. Zhu, T. Zhang, Y. Gou, Z. Xie, Y. Jin, Y. Wang, L. Song and M. Zhang, Environmental-friendly low-cost direct regeneration of cathode material from spent $LiFePO_4$, *J. Alloys Compd.*, 2022, **924**, 166612, DOI: **10.1016/j.jallcom.2022.166612**.

37 N. Hayagan, I. Gaalich, P. Loubet, L. Croguennec, C. Aymonier, G. Philippot and J. Olchowka, Challenges and Perspectives for Direct Recycling of Electrode Scraps and End-of-Life Lithium-ion Batteries, *Batteries Supercaps*, 2024, **7**, 2400120, DOI: **10.1002/batt.202400120**.

38 D. L. Thompson, J. M. Hartley, S. M. Lambert, M. Shiref, G. D. J. Harper, E. Kendrick, P. Anderson, K. S. Ryder, L. Gaines and A. P. Abbott, The importance of design in lithium ion battery recycling-a critical review, *Green Chem.*, 2020, **22**, 7585–7603, DOI: **10.1039/d0gc02745f**.

39 M. Muratori, M. Alexander, D. Arent, M. Bazilian, P. Cazzola, E. M. Dede, J. Farrell, C. Gearhart, D. Greene, A. Jenn, M. Keyser, T. Lipman, S. Narumanchi, A. Pesaran, R. Sioshansi, E. Suomalainen, G. Tal, K. Walkowicz and J. Ward, The rise of electric vehicles—2020 status and future expectations, *Prog. Energy*, 2021, **3**, 022002, DOI: **10.1088/2516-1083/abe0ad**.

40 M. Zhang, L. Wang, S. Wang, T. Ma, F. Jia and C. Zhan, A Critical Review on the Recycling Strategy of Lithium Iron Phosphate from Electric Vehicles, *Small Methods*, 2023, **7**, 1–14, DOI: **10.1002/smtd.202300125**.

41 S. Jiang, X. Li, Q. Gao, X. Lyu, S. N. Akanyange, T. Jiao and X. Zhu, Review on full-component green recycling of spent lithium iron phosphate cathode materials: From the perspective of economy and efficiency, *Sep. Purif. Technol.*, 2023, **324**, 124630, DOI: **10.1016/j.seppur.2023.124630**.

42 J. Harty, *Scrap battery supply forecast*, Fastmarkets Glob. Ltd, 2023. **https://www.fastmarkets.com/insights/six-key-trends-battery-recycling-market/** (accessed April 16, 2025).

43 H. Gao, D. Tran and Z. Chen, Seeking direct cathode regeneration for more efficient lithium-ion battery recycling, *Curr. Opin. Electrochem.*, 2022, **31**, 100875, DOI: **10.1016/j.coelec.2021.100875**.

44 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, *Adv. Funct. Mater.*, 2024, **34**, 2405055, DOI: **10.1002/adfm.202405055**.

45 X. Li, J. Zhang, D. Song, J. Song and L. Zhang, Direct regeneration of recycled cathode material mixture from scrapped $LiFePO_4$ batteries, *J. Power Sources*, 2017, **345**, 78–84, DOI: **10.1016/j.jpowsour.2017.01.118**.

46 J. Li, Y. Wang, L. Wang, B. Liu and H. Zhou, A facile recycling and regeneration process for spent $LiFePO_4$ batteries, *J. Mater. Sci.: Mater. Electron.*, 2019, **30**, 14580–14588, DOI: **10.1007/s10854-019-01830-y**.

47 B. Chen, M. Liu, S. Cao, G. Chen, X. Guo and X. Wang, Regeneration and performance of $LiFePO_4$ with Li_2CO_3 and $FePO_4$ as raw materials recovered from spent $LiFePO_4$ batteries, *Mater. Chem. Phys.*, 2022, **279**, 125750, DOI: **10.1016/j.matchemphys.2022.125750**.

48 K. Liu, S. Yang, F. Lai, Q. Li, H. Wang, T. Tao, D. Xiang and X. Zhang, Application of $H_4P_2O_7$ as leaching acid in one-

step selective recovery for metals from spent $LiFePO_4$ batteries, *Ionics*, 2021, **27**, 5127–5135, DOI: 10.1007/s11581-021-04273-w.

49 S.-H. Zheng, X.-T. Wang, Z.-Y. Gu, H.-Y. Lü, X.-Y. Zhang, J.-M. Cao, J.-Z. Guo, X.-T. Deng, Z.-T. Wu, R.-H. Zeng and X.-L. Wu, Direct and rapid regeneration of spent $LiFePO_4$ cathodes via a high-temperature shock strategy, *J. Power Sources*, 2023, **587**, 233697, DOI: 10.1016/j.jpowsour.2023.233697.

50 M. Fan, X. Chang, X. H. Meng, C. F. Gu, C. H. Zhang, Q. Meng, L. J. Wan and Y. G. Guo, Structural Restoration of Degraded $LiFePO_4$ Cathode with Enhanced Kinetics Using Residual Lithium in Spent Graphite Anodes, *CCS Chem.*, 2023, **5**, 1189–1201, DOI: 10.31635/ccschem.022.202201996.

51 J. Yang, K. Zhou, R. Gong, Q. Meng, Y. Zhang and P. Dong, Direct regeneration of spent $LiFePO_4$ materials via a green and economical one-step hydrothermal process, *J. Environ. Manage.*, 2023, **348**, 119384, DOI: 10.1016/j.jenvman.2023.119384.

52 B. Chen, M. Liu, S. Cao, H. Hu, G. Chen, X. Guo and X. Wang, Direct regeneration and performance of spent $LiFePO_4$ via a green efficient hydrothermal technique, *J. Alloys Compd.*, 2022, **924**, 166487, DOI: 10.1016/j.jallcom.2022.166487.

53 X. Tang, R. Wang, Y. Ren, J. Duan, J. Li and P. Li, Effective regeneration of scrapped $LiFePO_4$ material from spent lithium-ion batteries, *J. Mater. Sci.*, 2020, **55**, 13036–13048, DOI: 10.1007/s10853-020-04907-w.

54 Y. Xu, B. Zhang, Z. Ge, H. Wang, N. Hong, X. Xiao, B. Song, Y. Zhang, Y. Tian, W. Deng, G. Zou, H. Hou and X. Ji, Direct recovery of degraded $LiFePO_4$ cathode via mild chemical relithiation strategy, *Chem. Eng. J.*, 2023, **477**, 147201, DOI: 10.1016/j.cej.2023.147201.

55 X. Zhu, X. Ren, J. Chen, M. Gong, R. Mo, S. Luo and S. Yang, One-step regeneration and upgrading of spent $LiFePO_4$ cathodes with phytic acid, *Nanoscale*, 2024, **16**, 3417–3421, DOI: 10.1039/d3nr05387c.

56 Y. Yang, Z. Liu, J. Zhang, Y. Chen and C. Wang, Economical and low-carbon regeneration of spent $LiFePO_4$ materials by hydrothermal relithiation, *J. Alloys Compd.*, 2023, **947**, 169660, DOI: 10.1016/j.jallcom.2023.169660.

57 Q. Jing, J. Zhang, Y. Liu, W. Zhang, Y. Chen and C. Wang, Direct Regeneration of Spent $LiFePO_4$ Cathode Material by a Green and Efficient One-Step Hydrothermal Method, *ACS Sustainable Chem. Eng.*, 2020, **8**, 17622–17628, DOI: 10.1021/acssuschemeng.0c07166.

58 C. Wu, J. Hu, L. Ye, Z. Su, X. Fang, X. Zhu, L. Zhuang, X. Ai, H. Yang and J. Qian, Direct Regeneration of Spent Li-Ion Battery Cathodes via Chemical Relithiation Reaction, *ACS Sustainable Chem. Eng.*, 2021, **9**, 16384–16393, DOI: 10.1021/acssuschemeng.1c06278.

59 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*, 2023, **1**, 2341–2349, DOI: 10.1039/d3su00237c.

60 M. J. Ganter, B. J. Landi, C. W. Babbitt, A. Anctil and G. Gaustad, Cathode refunctionalization as a lithium ion battery recycling alternative, *J. Power Sources*, 2014, **256**, 274–280, DOI: 10.1016/j.jpowsour.2014.01.078.

61 T. Ouaneche, M. Courty, L. Stievano, L. Monconduit, C. Guéry, M. T. Sougrati and N. Recham, Room temperature efficient regeneration of spent $LiFePO_4$ by direct chemical lithiation, *J. Power Sources*, 2023, **579**, 233248, DOI: 10.1016/j.jpowsour.2023.233248.

62 T. Ouaneche, L. Stievano, L. Monconduit, C. Guéry, M. T. Sougrati and N. Recham, The art of Lithiation Revisited: Solvent-free room temperature reaction, *Energy Storage Mater.*, 2024, **70**, 103507, DOI: 10.1016/j.ensm.2024.103507.

63 X. Liu, M. Wang, L. Deng, Y. J. Cheng, J. Gao and Y. Xia, Direct Regeneration of Spent Lithium Iron Phosphate via a Low-Temperature Molten Salt Process Coupled with a Reductive Environment, *Ind. Eng. Chem. Res.*, 2022, **61**, 3831–3839, DOI: 10.1021/acs.iecr.1c05034.

64 K. Jia, J. Ma, J. Wang, Z. Liang, G. Ji, Z. Piao, R. Gao, Y. Zhu, Z. Zhuang, G. Zhou and H. M. Cheng, Long-Life Regenerated $LiFePO_4$ from Spent Cathode by Elevating the d-Band Center of Fe, *Adv. Mater.*, 2023, **35**, 2208034, DOI: 10.1002/adma.202208034.

65 C. D. Quilty, D. Wu, W. Li, D. C. Bock, L. Wang, L. M. Housel, A. Abraham, K. J. Takeuchi, A. C. Marschilok and E. S. Takeuchi, Electron and Ion Transport in Lithium and Lithium-Ion Battery Negative and Positive Composite Electrodes, *Chem. Rev.*, 2023, **123**, 1327–1363, DOI: 10.1021/acs.chemrev.2c00214.

66 Y. Li, L. Wang, K. Zhang, F. Liang, Y. Yao and L. Kong, High performance of $LiFePO_4$ with nitrogen and phosphorus dual-doped carbon layer for lithium-ion batteries, *J. Alloys Compd.*, 2022, **890**, 161617, DOI: 10.1016/j.jallcom.2021.161617.

67 C. R. Birkl, M. R. Roberts, E. McTurk, P. G. Bruce and D. A. Howey, Degradation diagnostics for lithium ion cells, *J. Power Sources*, 2017, **341**, 373–386, DOI: 10.1016/j.jpowsour.2016.12.011.

68 T. Yang, D. Luo, A. Yu and Z. Chen, Enabling Future Closed-Loop Recycling of Spent Lithium-Ion Batteries: Direct Cathode Regeneration, *Adv. Mater.*, 2023, **35**, 2203218, DOI: 10.1002/adma.202203218.

69 T. Wang, X. Yu, M. Fan, Q. Meng, Y. Xiao, Y. X. Yin, H. Li and Y. G. Guo, Direct regeneration of spent $LiFePO_4$ via a graphite prelithiation strategy, *Chem. Commun.*, 2019, **56**, 245–248, DOI: 10.1039/c9cc08155k.

70 H. Tian, M. Graczyk-Zajac, A. Kessler, A. Weidenkaff and R. Riedel, Recycling and Reusing of Graphite from Retired Lithium-ion Batteries: A Review, *Adv. Mater.*, 2024, **36**, 2308494, DOI: 10.1002/adma.202308494.

71 W. Mrozik, M. A. Rajaeifar, O. Heidrich and P. Christensen, Environmental impacts, pollution sources and pathways of spent lithium-ion batteries, *Energy Environ. Sci.*, 2021, **14**, 6099–6121, DOI: 10.1039/d1ee00691f.

72 C. Delacourt, C. Wurm, P. Reale, M. Morcrette and C. Masquelier, Low temperature preparation of optimized

phosphates for Li-battery applications, *Solid State Ionics*, 2004, **173**, 113–118, DOI: **10.1016/j.ssi.2004.07.061**.

73 G. Mali, C. Sirisopanaporn, C. Masquelier, D. Hanzel and R. Dominko, Li_2FeSiO_4 polymorphs probed by 6Li MAS NMR and 57Fe Mössbauer spectroscopy, *Chem. Mater.*, 2011, **23**, 2735–2744, DOI: **10.1021/cm103193a**.

74 I. F. Myronyuk, V. I. Mandzyuk, V. M. Sachko and V. M. Gun'ko, Structural Features of Carbons Produced Using Glucose, Lactose, and Saccharose, *Nanoscale Res. Lett.*, 2016, **11**, 508, DOI: **10.1186/s11671-016-1723-z**.

75 J. Chen, S. Wang and M. S. Whittingham, Hydrothermal synthesis of cathode materials, *J. Power Sources*, 2007, **174**, 442–448, DOI: **10.1016/j.jpowsour.2007.06.189**.

76 B. Gangaja, S. Nair and D. Santhanagopalan, Reuse, Recycle, and Regeneration of LiFePO 4 Cathode from Spent Lithium-Ion Batteries for Rechargeable Lithium- and Sodium-Ion Batteries, ACS Sustain, *Chem. Eng.*, 2021, **9**, 4711–4721, DOI: **10.1021/acssuschemeng.0c08487**.

77 L. Laffont, C. Delacourt, P. Gibot, M. Y. Wu, P. Kooyman, C. Masquelier and J. M. Tarascon, Study of the $LiFePO_4/FePO_4$ Two-Phase System by High-Resolution Electron Energy Loss Spectroscopy, *Chem. Mater.*, 2006, **18**, 5520–5529, DOI: **10.1021/cm0617182**.

78 D. Yang, Z. Fang, Y. Ji, Y. Yang, J. Hou, Z. Zhang, W. Du, X. Qi, Z. Zhu, R. Zhang, P. Hu, L. Qie and Y. Huang, A Room-Temperature Lithium-Restocking Strategy for the Direct Reuse of Degraded $LiFePO_4$ Electrodes, *Angew. Chem., Int. Ed.*, 2024, **63**, e202409929, DOI: **10.1002/anie.202409929**.

79 Y. Shi, M. Zhang, Y. S. Meng and Z. Chen, Ambient-Pressure Relithiation of Degraded $LixNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ ($0 < x < 1$) via Eutectic Solutions for Direct Regeneration of Lithium-Ion Battery Cathodes, *Adv. Energy Mater.*, 2019, **9**, 1900454, DOI: **10.1002/aenm.201900454**.

80 A. Örnek, E. Bulut and M. Özacar, The chemical, physical and electrochemical effects of carbon sources on the nano-scale $LiFePO_4$ cathode surface, *Ceram. Int.*, 2014, **40**, 15727–15736, DOI: **10.1016/j.ceramint.2014.07.096**.

81 X. Lv, J. Lin, X. Sun, Q. Huang, X. Zhang, T. Yu, E. Fan, Y. Ye, R. Chen, F. Wu and L. Li, Direct Recycling of Spent $LiFePO_4$ Cathodes Through Photocatalytic Correction of Anti-Site Defects, *Adv. Mater.*, 2025, 2503398, DOI: **10.1002/adma.202503398**.

82 H. S. Kim and E. J. Shin, Re-synthesis and electrochemical characteristics of $LiFePO_4$ cathode materials recycled from scrap electrodes, *Bull. Korean Chem. Soc.*, 2013, **34**, 851–855, DOI: **10.5012/bkcs.2013.34.3.851**.

83 D. Bian, Y. Sun, S. Li, Y. Tian, Z. Yang, X. Fan and W. Zhang, A novel process to recycle spent $LiFePO_4$ for synthesizing $LiFePO_4$/C hierarchical microflowers, *Electrochim. Acta*, 2016, **190**, 134–140, DOI: **10.1016/j.electacta.2015.12.114**.

84 L. Wang, J. Li, H. Zhou, Z. Huang, S. Tao, B. Zhai, L. Liu and L. Hu, Regeneration cathode material mixture from spent lithium iron phosphate batteries, *J. Mater. Sci.: Mater. Electron.*, 2018, **29**, 9283–9290, DOI: **10.1007/s10854-018-8958-7**.

85 P. Zhu, Z. Yang, H. Zhang, J. Yu, Z. Zhang, J. Cai and C. Li, Utilizing egg lecithin coating to improve the electrochemical performance of regenerated lithium iron phosphate, *J. Alloys Compd.*, 2018, **745**, 164–171, DOI: **10.1016/j.jallcom.2018.02.119**.

86 J. Zhou, X. Zhou, W. Yu, Z. Shang, Y. Yang and S. Xu, Solvothermal strategy for direct regeneration of high-performance cathode materials from spent lithium-ion battery, *Nano Energy*, 2024, **120**, 109145, DOI: **10.1016/j.nanoen.2023.109145**.

87 International Organization for Standardization 14044, Environmental Management, Life Cycle Assessment. Requirements and Guidelines, ISO 140442006, 2007, vol. 3, pp. 16.

88 A. Nordelöf, S. Poulikidou, M. Chordia, F. B. de Oliveira, J. Tivander and R. Arvidsson, Methodological approaches to end-of-life modelling in life cycle assessments of lithium-ion batteries, *Batteries*, 2019, **5**, 51, DOI: **10.3390/batteries5030051**.

89 M. Mohr, J. F. Peters, M. Baumann and M. Weil, Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes, *J. Ind. Ecol.*, 2020, **24**, 1310–1322, DOI: **10.1111/jiec.13021**.

90 T. Feng, W. Guo, Q. Li, Z. Meng and W. Liang, Life cycle assessment of lithium nickel cobalt manganese oxide batteries and lithium iron phosphate batteries for electric vehicles in China, *J. Energy Storage*, 2022, **52**, 104767, DOI: **10.1016/j.est.2022.104767**.

91 D. Weigl and D. Young, Impact of automated battery sorting for mineral recovery from lithium-ion battery recycling in the United States, *Resour., Conserv. Recycl.*, 2023, **192**, 106936, DOI: **10.1016/j.resconrec.2023.106936**.

92 M. Guerra, Can Automation Solve EV Battery Recycling Challenges?, *Battery Technol.*, 2024, 1–10.

93 M. Kessler, *Automation solution from Bosch Rexroth industrializes deep discharge of vehicle batteries*, Rexroth, A Bosch Co., 2024, pp. 1–3.

94 C. Stauch, *ReUse – Revolutionizing low-value LFP Battery Waste Recycling*, Fraunhofer ISC, 2024, pp. 7–9.

95 Q. Zhai, H. Jiang, N. Long, Q. Kang, X. Meng, M. Zhou, L. Yan and T. Ma, Machine learning for full lifecycle management of lithium-ion batteries, *Renewable Sustainable Energy Rev.*, 2024, **202**, 114647, DOI: **10.1016/j.rser.2024.114647**.

96 T. Zhao, W. Li, M. Traversy, Y. Choi, A. Ghahreman, Z. Zhao, C. Zhang, W. Zhao and Y. Song, A review on the recycling of spent lithium iron phosphate batteries, *J. Environ. Manage.*, 2024, **351**, 119670, DOI: **10.1016/j.jenvman.2023.119670**.

97 G. Harper, R. Sommerville, E. Kendrick, L. Driscoll, P. Slater, R. Stolkin, A. Walton, P. Christensen, O. Heidrich, S. Lambert, A. Abbott, K. Ryder, L. Gaines and P. Anderson, Recycling lithium-ion batteries from electric vehicles, *Nature*, 2019, **575**, 75–86, DOI: **10.1038/s41586-019-1682-5**.