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Recovery of $LiFePO_4$ cathodes: Criticalities and prospect towards a long-term eco-friendly solution

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ABSTRACT

Lithium iron phosphate batteries are currently getting increasing attention due to their low cost, good cycling stability and high safety characteristics. Enhanced batteries consumption is accompanied by their subsequent discard that, if poorly managed, may cause serious environmental damage as well as severe economic losses. Numerous cathode recycling techniques have been developed, including pyro/hydrometallurgical methods and direct regeneration.

Besides providing information on the working and failing mechanism of these batteries, this review aims to present the most relevant spent cathode treatments, with an emphasis on environmental and economic implications. Moreover, it examines global regulations on lithium-ion battery recycling and identifies unresolved issues that still need to be addressed.

Our analysis revealed that using organic acids significantly enhances the economic feasibility of hydrometallurgical processes, enabling for selective lithium recovery. This approach not only simplifies the procedure but also makes it more eco-friendly. Direct regeneration techniques, on the other hand, emerge as both economical and green alternatives to produce new cathodes with excellent electrochemical properties, but are difficult to apply due to the variability of the spent materials to be treated. Lastly, it appears imperative for legislators to formulate a regulatory framework specifically tailored to the treatment of spent lithium-ion batteries.

1. Introduction

Driven by the need to cut greenhouse gas emissions and address the depletion of fossil fuels, the adoption of clean technologies has surged at an unprecedented rate in recent years. According to the International Energy Agency (IEA), between 2015 and 2022, installed solar photovoltaic capacity increased by 400% while electric car sales grew by 2000%, with over 25 million units sold [1]. These trends were accompanied by an increasing need for energy storage devices, which pushed lithium-ion batteries (LIBs) to the forefront due to their features such as high energy density, low self-discharge, and long cycle life [2].

LIBs can be classified based on the crystal structure of their cathodes into three types: spinel, layered, and polyanion (Fig. 1). Layered cathodes, such as $LiCoO_2$ (LCO) or $LiNi_xCo_yMn_zO_2$ (NCM), are the most widespread due to their high energy density but are losing popularity due to the high cost of cobalt (Co) and their toxicity. Spinel compounds are represented by the $LiMn_2O_4$ (LMO) cathode, which, although quite economical, is not widely used due to its low specific power and short life [3]. Lastly, polyanionic compounds, particularly $LiFePO_4$ (LFP) cathodes, have gained increasing success in recent years, despite their relatively low energy density. This tendency is explained by their low cost, as components like iron (Fe) and phosphorus (P) are abundant in nature, and by their long life and high safety due to the strong Fe-O and P-O bonds which make their olivine structure extremely stable [4]. Particularly, driven by the preference of Chinese electric vehicles (EVs) manufacturers, in 2022 lithium iron phosphate batteries reached their highest market share of the last decade, settling just below 30 % [5].

It is natural to assume that such a significant increase in LFP batteries sales will go hand in hand with an increase in their scrapping and, as their average life is 5–10 years, this will soon become a global issue. Failure to properly dispose of these devices would not only cause serious pollution problems due to the release of toxic compounds into the environment but would also lead to the loss of strategic elements such as lithium and phosphorus, which are considered critical raw materials by the European Union [7]. However, conventional recycling techniques for spent batteries are not as profitable as expected when applied to LFP

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cathodes due to the lower value of lithium and iron compared to nickel, cobalt and manganese. Researchers are therefore focusing on the development of shorter and less chemical-intensive processes and the direct regeneration of such materials [8].

Rather than describing in detail all the work done on LFP recycling, a subject already covered by different existing reviews [3,4,8–12], this paper aims to summarise in one concise piece of work relevant information on the functioning and fading mechanism of LFP batteries, on LIBs recycling policies, on the most consolidated techniques for recovering the cathode material, and last, but not least, on the most recent and environmentally friendly methods proposed in the literature, such as those based on the use of biocompatible organic materials as leaching agents and mechanochemical oxidation techniques. The work is therefore intended as a useful introduction of the LIB's issue for a large, even non expert, audience.

2. Functioning and failure of $LiFePO_4$ batteries

Typically, LIBs consist of several key elements: anode and cathode plates, a separator and a liquid electrolyte, all enclosed in a sealed shell (Fig. 2). The cathode is made of an aluminium foil coated with the active material. Similarly, the anode consists of a copper sheet coated with a graphite layer. The use of a binder, usually polyvinylidene fluoride (PVDF), ensures proper adherence between electrodes components. The electrolyte generally comprises inorganic lithium salts, usually hexafluorophosphate ($LiPF_6$), dissolved in an organic solvent, such as ethylene carbonate, diethyl carbonate, dimethyl carbonate, or a mixture thereof. To mitigate the risk of short circuits between the electrodes, these are kept segregated by a separator, consisting of a Li^+ permeable polymeric membrane [10]. Fig. 2 also shows a detail of the crystal structure of the cathode, where it can be seen that FeO_6 occupies the octahedron sites and PO_4 occupies the tetrahedron sites [13].

The good cycling performance of LFP batteries is inextricably linked to their stable olivine structure. In fact, during charging, LFP gradually releases lithium ions to form heterosite ($FePO_4$), and during discharging, lithium ions insert into iron phosphate (FP) to yield triphylite again (Equation (1)), without significantly altering the framework [9]. The extremely stable structure of these cathodes also constitutes a sort of limitation, as Li^+ ions can only diffuse along one-dimensional channels, resulting in poor rate capability [14]. Commercially available cathodes are usually modified using conductive additives to enhance their kinetics, with LFP/carbon black composites standing out for their remarkable conductivity and ease of production [15].

$$LiFePO_4 - xLi^+ - xe^- \leftrightarrow xFePO_4 + (1-x)LiFePO_4 \tag{1}$$

Over time, LIBs undergo a deterioration in both their electrochemical and structural properties. Once LIBs have reached 80% of their original capacity, it is common to consider them as exhausted. Although the decline in capacity is often attributed to the cathode, it is caused by inefficiencies in other battery components. The main reason for capacity loss is the depletion of available Li^+ ions, which mainly occurs at the anode due to the formation of the solid electrolyte interface (SEI) [8]. Such interface, formed by redox reactions taking place at the anode-electrolyte boundary and involving solvated Li^+, consists of a thin solid film deposited on the surface of the electrode [16]. Being an electronically insulating but ionic conducting passivation layer, the SEI is beneficial to improve the self-discharge characteristics, cycle life, rate capability and safety of the battery [17]. Indeed, it allows Li^+ to reach the negative electrode, while protecting it from electrolyte anions. Anyhow, due to structural and chemical factors, after its first deposition, SEI may continue to thicken or be destructed and reformed during prolonged charge-discharge cycles, resulting in the progressive loss of active material [18]. The presence of a passivation layer, similar to the SEI, on the surface of the positive electrode has also been reported [19]. This film, which takes the name of cathode electrolyte interface (CEI) is usually thinner than SEI, yet it is detrimental to the battery performance as it increases the ionic resistance and accelerates the degradation of the active carbon coating [20].

Lithium deficiency is directly related to the structural degradation of the LFP cathode [21]. Li^+ vacancies induce the migration of Fe^{2+} to the Li site to form "anti-site" defects, which hinder ionic diffusion, thereby limiting the rate performance of the battery [22]. The structural integrity of the cathode is further compromised by the phase transition between LFP and FP during operation. Indeed, the intercalation and deintercalation of Li^+, generates a biphasic material with sharp interfaces between the constituents [23]. This phase transition, accompanied by a 6.8% volume change [24], generates stress along the interface leading to the formation of pores and fractures inside the polyanionic structure [25]. Repeated cycling favours the nucleation of such cracks leading to the loss of electrical contact between active material grains and subsequent degradation of battery capacity.

Additionally, electrolyte corrosion is another factor contributing to the aging of LFP batteries. This phenomenon, enhanced by high operating temperatures, consists of the dissolution of the iron present in the cathode as a consequence of the attack of hydrofluoric acid generated by the hydrolysis of the electrolyte [26]. During charging, solvated Fe^{2+} migrates to the anode and is therein reduced [27]. The iron thus formed is deposited on the graphite layer, making it less available for Li ions insertion and favouring the thickening of the SEI [28].

3. $LiFePO_4$ cathode recycling

Although not the major factor causing battery degradation, attention

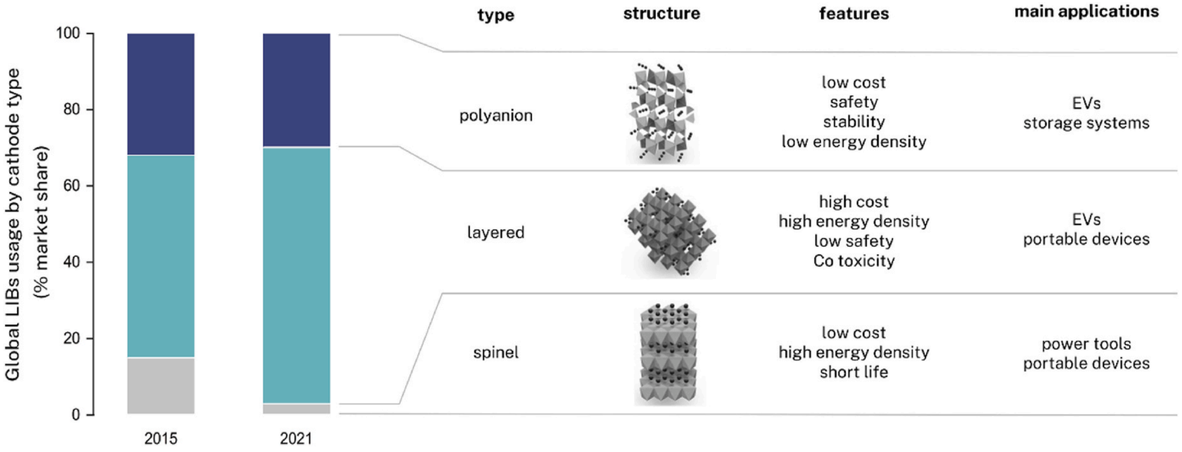

Fig. 1. Market share for different types of LIBs cathodes [6] and comprehensive comparison among them.

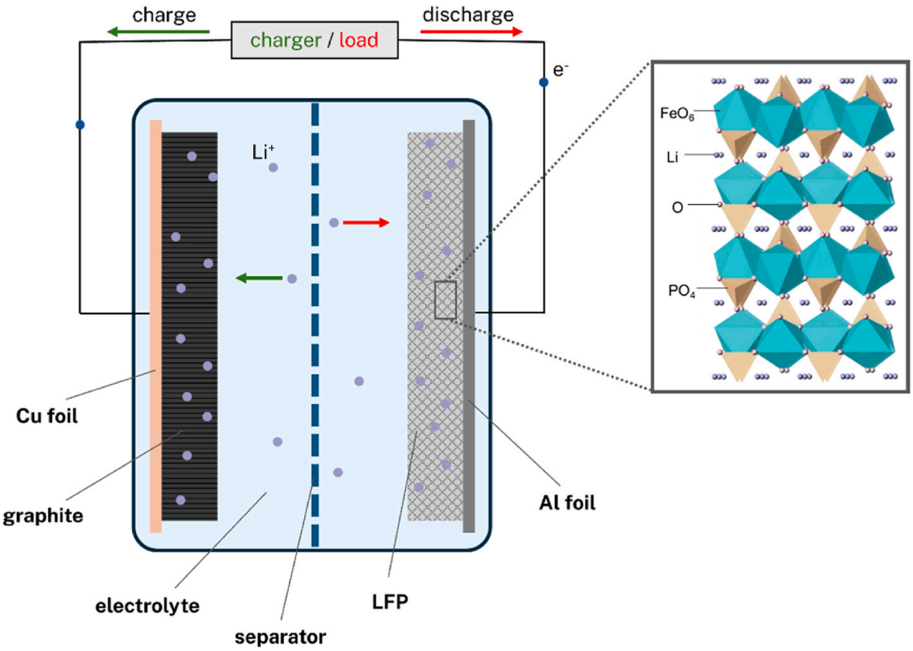

Fig. 2. The composition of LFP battery cell and its olivine crystalline frame.

is focused on recycling LFP cathodes due to their valuable materials content and production complexity.

As shown in Fig. 3, the initial steps to obtain spent cathode powder involve pre-treating retired batteries through discharging, dismantling, and separation. Proper application of the pretreatment process is crucial, as it simplifies subsequent procedures and guarantees operational safety.

Discharging is essential to prevent short-circuiting during recycling, thereby mitigating the risk of fire or explosion. There are two primary methods of discharging: passive discharge, where the waste LFP battery is connected to an external discharge device to neutralize the residual charge [40], and self-discharge, where the battery is immersed in a salt solution to release the stored energy.

Disassembly has the aim to separate and recycle the various components of the LFP battery [41]. Disassembly can either be performed manually or mechanically. Manual disassembly allows for the complete recovery of components such as plastic casings, diaphragms, copper foil, and aluminum foil. However, being a cumbersome and low-efficiency process, manual disassembly is typically reserved for small-scale processing in laboratory research [42]. Conversely, mechanical disassembly is predominantly utilized on the industial scale due to its enhanced productivity and reduced costs. In this method, waste LFP batteries are generally shredded in their entirety [43]. Following the shredding process, additional separation steps are needed to retrieve the active material.

Once the active material is obtained, it is either recovered to restore its electrochemical properties or separated into its components, as will be discussed in the following sections.

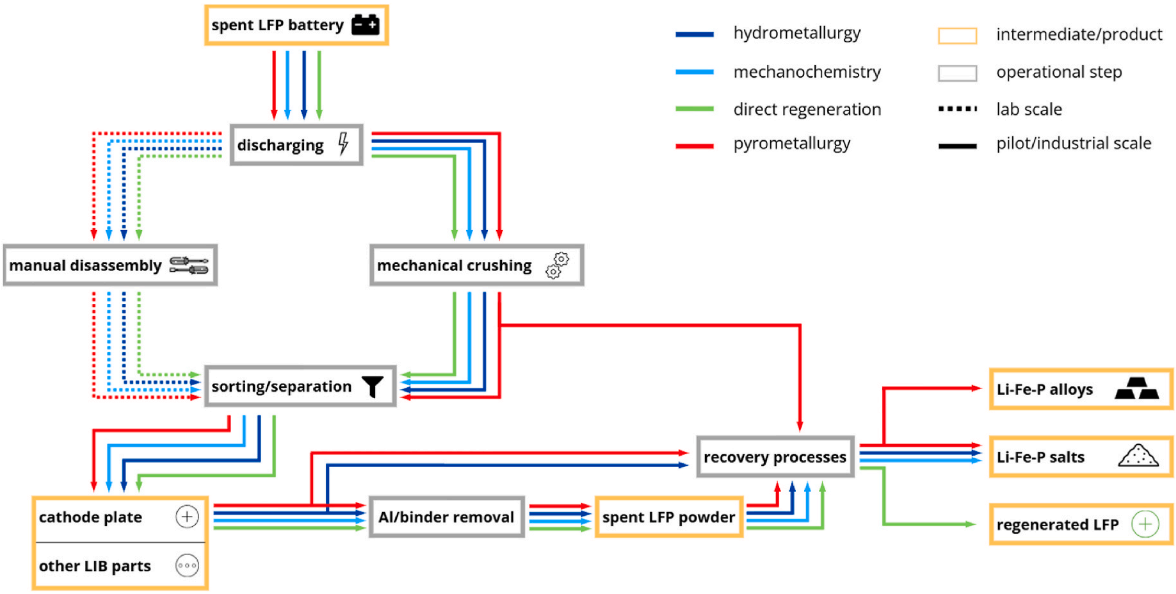

Fig. 3. Schematic representation of possible spent LFP recycling routes.

3.1. Metal recovery

The need for metals is growing in today's society and is predicted to do so in the near future due to factors like urbanization, the construction of new infrastructure in developing nations, the widespread use of electronics, and advancements in energy technologies. Hence, metals are considered critical raw materials as they are of the highest importance for the economy [44]. In this context, metal recovery is a good example of an integrated approach, since, when compared to the conventional production process starting from ores, it requires at least two orders of magnitude less energy per kilogram of produced metal [45]. Metals are almost endlessly recyclable as they do not degrade. On the other hand, their dispersion in nature can lead to a significant harm for environment and human health. Thus, on the long term, a sustainable approach to managing metals requires closing cycles as much as feasible. From an industrial application standpoint, it is important to balance considerations of low cost, high efficiency, high product value and environmental impact [46].

Following are the main techniques for recovering metals from spent LFP battery cathodes, including pyrometallurgy, hydrometallurgy and mechanochemistry.

3.1.1. Pyrometallurgy

Pyrometallurgy is a process that uses high temperatures to cause physical and chemical changes that allow for metal recovery [11]. It was traditionally used to extract valuable metals from ores but has become the go-to technology in the LIBs recycling industry due to its operational simplicity [47]. The extractive pyrometallurgical options used to process spent lithium-ion batteries are roasting/calcination and smelting [48], which differ mainly in operating temperature and atmosphere. Zhang et al. analysed LFP roasting in a sulphatising atmosphere [49]. Results showed that a mild temperature (600 °C) reaction with sodium-bisulphate can convert lithium inside the olivine structure of the cathode into water-soluble sulphate, leaving Fe-P-O containing species in the solid state. Carbothermic reduction (CTR) has also been investigated as a Li recovery procedure [50]. Researchers exploited sodium carbonate (Na_2CO_3) action to break down the olivine structure of LFP so that C can reduce iron oxides to metallic iron, which, after a 20-min water leaching at room temperature, was separated from the Li^+ containing solution by magnetic separation. Interestingly, the carbon source used for the CTR was the graphite contained in the anode.

Being capital-intensive due to high energy requirements and complicated off-gas treatment operations, pyrometallurgy is well suited to treating cathodes containing precious metals such as NMC and LCO rather than the relatively inexpensive LFP for which it results unprofitable. In fact, although pyrometallurgical processes are currently the only industrially established treatments, they struggle to generate economic returns when applied to LFP cathodes due to the low added value of the recovered products. This is in contrast to LCO cathodes, which contain more valuable materials. Furthermore, its application to the LFP powders is limited by its reduced ability to recover lithium [51], which easily moves to the gas phase at high temperatures.

The economic performance of pyrometallurgical processes, as well as their environmental one, can be enhanced reducing their operating temperature. It is the case of the NaOH-assisted roasting proposed by Li et al. [52], in which the metals contained in the spent cathode were recovered at 150 °C. In this process, the oxidizing action of sodium hydroxide on Fe was exploited to break the LFP stable olivine structure, allowing the release of Li which was recovered in the form of highly pure lithium phosphate (Li_3PO_4) through subsequent water leaching and precipitation.

3.1.2. Hydrometallurgy

With the advantages of high metal recovery and reduced energy consumption, hydrometallurgical techniques are the preferred ones for spent LFP treatment. The typical hydrometallurgical process consists of a leaching step, aimed at dissolving part or all the components of the cathode in an aqueous medium, followed by a purification stage. Based on the reagent used to solubilize the cathode, these processes can be categorized into inorganic acid leaching, organic acid leaching and acid-free leaching (Table 1).

In the framework of inorganic acid leaching, sulphuric acid (H_2SO_4) is the most common agent due to its large availability and low cost [53]. The typical process exploiting sulphuric acid can effectively leach Li and Fe out of the spent cathode at relatively low temperature according to Eq. (2) [54]. Anyhow, being characterized by large acid consumption and non-selective extraction, it requires the adoption of complicated metals separation steps and onerous wastewater treatments. The use of oxidizing agents or appropriate pre-treatment can alleviate these problems [55]. Addition of hydrogen peroxide (H_2O_2) allowed Li et al. [56] to leach over 98% of Li, leaving Fe and P in the solid phase, using an almost stoichiometric H_2SO_4 amount (Eq. (3)), with obvious economic and environmental benefits. Tao et al. [57] were able to achieve such a result by preceding the inorganic acid reaction with an oxidative activation step at a temperature of 600 °C that removed the impurities covering the spent cathode powder and destroyed its stable olivine structure, thus facilitating lithium solubilization. Phosphoric acid [58], pyrophosphoric acid [59] and hydrochloric acid [60] are other examples of inorganic leaching agents that researchers have investigated.

$$2LiFePO_4 + 3H_2SO_4 \rightarrow 2FeSO_4 + Li_2SO_4 + 2H_3PO_4 \tag{2}$$

$$2LiFePO_4 + H_2SO_4 + H_2O_2 \rightarrow 2FePO_4\downarrow + Li_2SO_4 + 2H_2O \tag{3}$$

In view of the environmental risks associated with the use of mineral lixiviants, organic acids have become a major focus of research in the last few years [61]. This trend can be explained by the fact that, in addition to providing metal leaching efficiencies comparable to those of inorganic acids, these substances offer several advantages such as biocompatibility, mild operating conditions and facile downstream recovery [62]. Zhou et al. [63] proposed a closed-loop process exploiting the synergistic interaction between malic acid and hydrogen peroxide. In their study they found that Li extraction occurred due to an in-situ reaction directly converting the olivine structure of LFP to the olivine structure of FP. Under the optimized operating conditions reported in Table 1, researchers were able to leach 99.12% of the lithium present in the spent cathode with an outstanding selectivity. Precipitation with sodium carbonate (Na_2CO_3) was used to recover extracted Li cations as high purity lithium carbonate (Li_2CO_3), which was used, together with the previously obtained iron phosphate, to resynthesize LFP powder that showed electrochemical properties comparable to the fresh one. Lastly, following the principle of treating waste with waste, they used the small amount of Fe present in the leachate to effectively remove Arsenic from water. In the work of Yang et al. [64], relatively cheap acetic acid was used, together with H_2O_2, to achieve Li recovery. By using the cathode plate directly as starting material rather than the previously scratched powder, researchers were able to simplify the recycling process, demonstrating the high selectivity of organic lixiviants towards Al impurities, otherwise impossible to achieve using conventional inorganic acids. Leaching with formic acid/H_2O_2 systems was explored by Mahandra and Ghahreman [65]. In their research highly selective lithium extraction was followed by alkaline precipitation with sodium phosphate (Na_3PO_4) which allowed for the obtainment of battery grade lithium phosphate (Li_3PO_4). An oxidant free organic acid treatment was proposed by Chai et al. [66]. Using oxalic acid alone, more than 99% of the lithium present in the spent cathode powder, with only 2.4% Fe moving to the liquid phase, was extracted through a reaction carried out at 90 °C for 60 min. The researchers emphasised the ability of oxalic acid to act as both a proton donor and a reducing agent, promoting the release of Li^+ ions from the cathode matrix and providing oxalate ions for precipitating iron as ferrous oxalate at the same time, thus enabling a selective leaching process despite the absence of hydrogen peroxide. In the work of Jin et al. [67], oxidation was carried out by bubbling air into an aqueous solution with the cathode dispersed in it. O_2 has emerged as an excellent green oxidant even at room temperature, but the proposed process cannot be regarded as completely eco-friendly as it requires the addition of small quantities of highly concentrated sulphuric acid solutions to maintain the pH of the system at the desired level. Attempts to avoid using H_2O_2 are noteworthy because, although it only gives off oxygen and water as decomposition products, it cannot be considered a green reagent due to its energy-intensive production process [68]. Kumar et al. [69] investigated the possibility of using citrus fruit juices as leaching agents. Lemon juice, paired with hydrogen peroxide, stood out as an excellent reagent owing to its high citric acid content, which provides a suitable pH for selective lithium extraction.

In an attempt to find new solutions to achieve satisfactory metal recovery while maintaining mild operating conditions, different acid-free leaching processes have recently been proposed. Qiu et al. [70] achieved selective Li leaching by exploiting the oxidative potential of H_2O_2. Despite the advantages of short reaction time and almost neutral operating pH, their process has the drawbacks of excessive hydrogen peroxide consumption and reduced solid to liquid ratio which limit its scale-up. Zhang et al. [71] used sodium persulphate ($Na_2S_2O_8$) as both oxidizing and leaching agent, according to the reaction mechanism described by Eq. (4). Their study demonstrated that a small excess in $Na_2S_2O_8$ dosage resulted in a slightly acidic environment which turned out to be beneficial to selective Li extraction. The sustainability of the process was further enhanced by the possibility of downstream regeneration of the sodium salt. Similarly, Peng et al. [72] exploited ammonium persulphate ($(NH_4)_2S_2O_8$) to realize a “feed three birds with one scone” process which, being carried out on the cathode plate, allowed the simultaneous recovery of the metals composing the spent powder and of the aluminium support.

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$$2LiFePO_4 + Na_2S_2O_8 \rightarrow 2FePO_4\downarrow + Li_2SO_4 + 2Na_2SO_4 \tag{4}$$

Common oxidant sodium hypochlorite (NaClO) was used by Liu et al. [73] to avoid the use of acids. Results showed that the highly alkaline environment provided by the NaClO solution enhanced the precipitation of $Fe(OH)_3$, whose leaching turned out to be negligible. Ferric hydroxide was converted into Fe_2O_3, through calcination, and used with the recovered Li_3PO_4 to resynthesize high performance $LiFePO_4/C$. Isomorphic substitution between ferric and ferrous iron was explored as Li extraction technique by Niu et al. [74]. In their work, inorganic $FeCl_3$ salt was used to directly substitute Fe^{2+} in spent LFP, to form FP thus inducing Li^+ solubilization. Lithium was eventually recovered from the leachate by means of organic solvent extraction. Dai et al. [75] proposed two possible reactive systems involving the use of ferric sulphate ($Fe_2(SO_4)_3$). Their study proves the effectiveness of the non-oxidizing iron salt in performing the isomorphic substitution when employed in the theoretical molar ratio $Fe_2(SO_4)_3$:$LiFePO_4$ = 1:2. Further analysis showed the addition of reduced amounts of H_2O_2 was beneficial to the oxidation of Fe^{2+} back to Fe^{3+}, thus reducing salt consumption to one third of the theoretical molar amount.

Coupling hydrometallurgy to other processes could represent a feasible way to enhance the economic and environmental performance of metal recovery. It is the case of Xu et al. [76] work, which explores a combination between carbon dioxide (CO_2) sequestration and lithium extraction. Using the acidic environment provided by CO_2 dissolution in water and the action of H_2O_2, researchers developed a process capable of recovering 96.5% of the lithium present in the spent cathode. Having the ability to fix about 120 kg of CO_2 per tonne of processed LFP, this treatment represents a pioneering and highly green approach to recovering precious metals from spent LIBs. Mahandra et al. [77] used the neutrophilic bacterium Thiobacillus thioparus to selectively extract lithium from spent cathode powders. Thanks to the sulphuric acid produced during the bacterial metabolic activity, the proposed system allowed for the recovery of high purity Li salts from the leachate which was subsequently used as a culture medium to grow alkaliphilic bacteria. Despite the eco-friendliness of the proposed approach, further studies focused on processing time reduction are needed.

3.1.3. *Mechanochemistry*

Impact, high shear, compression, friction and extrusion are the means by which mechanochemistry is capable of reducing activation energies, thereby increasing reaction rates [78]. Mechanically induced alterations in LFP chemical and physical properties facilitate metal recovery from spent cathode powders, allowing for effective recycling under mild conditions.

Table 1
Hydrometallurgical processes for LFP cathodes recovery.

Leaching agent	Reactants	Operating conditions	Leaching efficiency	Ref.
Inorganic acid	H_2SO_4	H_2SO_4 2.5 M, S/L = 100 g/L, 4 h, 60 °C	Li 97% Fe 98%	[54]
	$H_2SO_4 + H_2O_2$	H_2SO_4 0.3 M, $n(H_2O_2/Li)$ = 2.07, 2 h, 60 °C	Li 96.85% P 1.95% Fe 0.027%	[56]
	HCl + NaClO	n(HCl/Li) = 0.8, n(NaClO/Li) = 1, 1 h, 50 °C	Li 99% Fe < 2%	[58]
	H_3PO_4 + Citric acid	H_3PO_4 2.3 M, $C_6H_8O_7$ 0.58M, S/L = 200 g/L 3h, 50 °C	Li 95.1% Fe 95.3% P 96.2%	[60]
Organic acid	Malic acid + H_2O_2	$n(C_4H_6O_5/Li)$ = 0.435, 4% vol. H_2O_2, S/L = 100 g/L, 0.5 h, 25 °C	Li 99.12% Fe < 1%	[63]
	Acetic acid + H_2O_2	CH_3COOH 0.8 M 6% vol. H_2O_2, S/L = 120 g/L, 0.5 h, 50 °C	Li 95.05% Fe 0.93%	[64]
	Formic acid + H_2O_2	HCOOH 1 M 5% vol. H_2O_2, S/L = 100 g/L, 0.5 h, 30 °C	Li 97.92% Fe < 0.5%	[65]
	Oxalic acid	$H_2C_2O_4$ 0.32 M S/L = 100 g/L, 1 h, 90 °C	Li 99.15% Fe 2.4%	[66]
	Lemon juice + H_2O_2	100% Lemon juice 6% vol. H_2O_2, S/L = 67 g/L, 1.5 h, room temperature	Li 94.83% Fe 4.05% P 0.84%	[69]
Acid free	H_2O_2	15% vol. H_2O_2, S/L = 10 g/L, 0.5 h, 50 °C	Li 97.6% Fe < 1%	[70]
	Air	Q_{AIR} = 600 mL/min S/L = 100 g/L, 5 h, 25 °C	Li 99.3% Fe/P < 0.02%	[67]
	NaClO	NaClO 0.54 M 1 h, 60 °C	Li 99.8% Fe 0.003%	[73]
	$FeCl_3$	$n(FeCl_3/LFP)$ = 1.3, S/L = 200 g/L, 0.5 h, 40 °C	Li 99% Fe 2%	[74]

Co-grinding with inexpensive and non-toxic sodium chloride (NaCl) was exploited by Liu et al. [79] to induce isomorphic substitution between sodium and lithium from scrapped LFP. Their study confirmed that similarities in Na and Li outer electron arrangements and coordination environments favoured the former over Fe as a lithium replacement during mechanically induced local atomic rearrangements. Water soluble lithium chloride (LiCl) was recovered through a 30-min dissolution of the ball-milled powder in deionized water, which also yielded a sodium-iron-phosphate precipitate. Reaction with Na_2CO_3 and pH adjustment allowed to obtain battery grade Li_2CO_3 and regenerated NaCl, thus achieving loop closure. To overcome the limitations related to the long reaction times needed to achieve the isomorphic substitution, the same research group proposed solid-state oxidation of $LiFePO_4$ with sodium persulfate [80]. Surprisingly, they were able to achieve the selective release of 99.7% of the Li contained in the olivine structure of the cathode through a 5-min ball milling at 600 rpm. With the advantages of reduced chemical consumption and fast reaction time, this process appears as a promising eco-friendly way to recover lithium from exhausted cathodes. Yang et al. [81] demonstrated how prior mechanochemical activation with chelating agent ethylenediaminetetraacetic acid disodium salt (EDTA-2Na) could improve the ability of diluted phosphoric acid solutions to extract metals from LFP powders. Indeed, Li and Fe leaching efficiencies increased from 61.7% and 37.8%, respectively, to 97.67% and 94.29% when the liquid phase reaction was preceded by ball milling for 2h at 550 rpm. The proposed process, however, has the drawbacks of inorganic acid usage and non-selective leaching, which imply complicated downstream processing. Liu et al. [82] proposed an ultra-fast mechanochemical reaction with EDTA-2Na in which the addition of H_2O_2, acting as both dispersing and oxidizing agent, allowed to achieve selective Li extraction while avoiding H_3PO_4 utilization. The presented strategy can immediately achieve effective de-intercalation of Li within 4 min while maintaining the olivine structure of $FePO_4$. Mechanically induced de-lithiation with oxalic acid was explored by Fan et al. [83]. Their process, which involved impurities removal through high temperature calcination, wet mechanochemical reaction and room temperature water leaching, selectively extracted 99% of lithium while simultaneously recovering more than 94% of Fe in the form of ferrous oxalate. Throughout their studies, researchers discovered that the addition of small amounts of water during ball milling increased the extraction capabilities of the system by eliminating dead angles and ensuring proper homogenisation. Li et al. [84] proposed citric acid as LFP co-grinding agent. Results showed that using water as aiding agent in place of hydrogen peroxide, induces a shift in the reaction mechanism leading to non-selective leaching. Indeed, without the oxidizing effect of H_2O_2, both Li and Fe, which are eventually found as citrates in the downstream leachate, are released during the mechanochemical reaction.

3.2. *Direct regeneration*

Usually involving expensive chemicals or energy-intensive processes, metal recovery may result unprofitable when applied to LFP cathodes. For this reason, many direct regeneration methods, targeting the main cathode degradation mechanisms, have recently been proposed. As already mentioned in section 2, Li loss is the primary cause of LFP batteries electrochemical decay, hence re-lithiation is the first step for proper regeneration. Moreover, spent cathodes are characterised by particle cracking and anti-site defects that require elements replenishment and energy supply to be addressed [85].

So far, high-temperature solid-state regeneration has been the most studied technique due to its efficiency and scalability [86–89]. Zou et al. [87] effectively restored spent LFP materials through high-temperature roasting in inert atmosphere, using Li_2CO_3 as lithium source and ascorbic acid as both reducing agent and carbon source. In their paper, researchers highlighted the importance to ensure stable electrochemical performance of the regenerated cathode of performing, prior to solid-state sintering, a high-temperature oxidation treatment to remove binder and residual electrolyte. Supplementation of Fe and P salts, together with lithium and carbon sources, allowed Liang et al. [89] to recover the lattice structure of the cathode together with its electrochemical properties in a single step process. Similarly, simultaneous structural and compositional regeneration was achieved by Ji et al. [90] using 3,4-dihydroxybenzonitrile dilithium (Li_2DHBN). Researchers found that the pyrolysis of the multifunctional organic salt allowed Li^+ ions to penetrate the $FePO_4$ matrix while preventing Fe(III) formation due to the reductive environment created by nitrile groups. Cathode repairing was eventually completed by the amorphous carbon formed during the process which covered the surface of the LFP particles.

Although highly effective, solid phase methods require substantial amounts of heat to be supplied to the reactive system to overcome the activation energy of regeneration reactions. For this reason, hydrothermal processes exploiting water as heat and mass transferor have been proposed. Chen et al. [91] proposed a simple and economic strategy for solution recrystallization using tartaric acid and lithium hydroxide. The regeneration process, involving 3h reaction at 200 °C and subsequent calcination in Ar atmosphere, allowed researchers to obtain a repaired cathode exhibiting capacity retention as high as 99.1% after 200 cycles at 1 C. Hydrothermal re-lithiation with hydrazine hydrate ($N_2H_4 \bullet H_2O$) and lithium sulphate (Li_2SO_4) was explored by Jing et al. [92]. In the proposed method, Li replenishment is achieved through a redox mechanism in which $N_2H_4 \bullet H_2O$ acts both as electron donor, to sustain the reaction, and as proton acceptor, to maintain a slightly alkaline environment beneficial to LFP.

Molten salt thermochemistry was used by Liu et al. [93] to achieve low temperature (300 °C) re-lithiation. Enhanced mass-transport phenomena in the molten lithium nitrate ($LiNO_3$) solution allowed for rapid insertion of lithium into the spent olivine structure, resulting in a regenerated cathode with better rate performance than pristine LFP at 5 C.

An electrochemical regeneration approach, resembling battery discharge, was proposed by Zhou et al. [94]. In their work, the authors developed a H-type electrolytic bath in which spent LFP powders acting as the cathode and a commercial zinc (Zn) plate acting as the anode, were separated by an anion-exchange membrane. Under the application of a small discharge current (5 mA), Li^+ ions contained in the electrolyte (lithium salt aqueous solution) were inserted into the scrapped cathode, according to Eq. (5).

$$x/2\,Zn + Li_{1-x}FePO_4 + xLi^+ \leftrightarrow LiFePO_4 + x/2\,Zn^{2+} \tag{5}$$

The use of pre-lithiated materials to regenerate spent cathode powders constitutes a clever example of advanced LFP treatment. Wang et al. [95] proposed the use of a Li-containing graphite anode to achieve lithium replenishment through the usual discharge half-cell reaction. Their process, although effective and innovative, leaves unaddressed the issues associated with the complexity of the pre-treatment. Fan et al. [96], on the other hand, significantly simplified the regeneration process replacing the conventional separator with a functional pre-lithium one. Such element, together with fresh graphite anode and spent LFP, was assembled into a new battery that, after one activation cycle, showed substantial capacity recovery.

Metal ions doping coupled to high-temperature solid-state regeneration has also been reported. Liu et al. [97] elevated the electrochemical performance of the regenerated cathode adding a vanadium (V) source during solid-state sintering. Researchers found that V insertion in some of the Fe sites reduced the volume of the LFP cell, resulting in increased Li^+ diffusivity coefficient. Similarly, nitrogen doping was exploited by Jia et al. [98] to obtain a long-lasting regenerated cathode via hydrothermal route.

Overall, all regenerated materials showed good performance. Zou et al. [87] obtained a material with excellent structural stability, able to cycle to capacitance values close to the theoretical ones (94%) and maintain 80% of the capacity after 800 cycles @ 1.0 C. The material regenerated by Chen et al. [91] also showed similar performances, with an initial specific capacitance equal to 89% of the theoretical one and a capacity retention of 99% after 200 cycles @ 1.0 C. Slightly lower were the results obtained by Jing et al. [92], with an initial specific capacitance equal to 83% of the theoretical one and a capacity retention higher than 98% after 200 cycles @ 1.0 C. Liu et al. [93] obtained a capacitance equal to 85% of the theoretical one with a capacity retention of 85% after 300 cycles @ 1.0 C. Zhou et al. [94] obtained a material with performances very similar to that prepared by Liu et al. with an initial performance just below 80% that retained 85% of the capacity after 300 cycles @ 1.0 C. Similarly, the material prepared by Liu et al. [97] at high temperature and doping with vanadium showed an initial capacity just below 80%, but showed exceptional cyclability: the capacity retention was 99% after 300 cycles @ 1.0 C.

Life cycle assessment of direct regeneration processes shows that their energy requirement is only 20% and 10% of that of pyrometallurgy and hydrometallurgy, respectively [99]. Moreover, their cost is only about 20% of the LFP cost, so they are promising from the economic standpoint [93]. However, technical challenges related to the presence of Al and Cu impurities deriving from spent LIBs dismantling and to the varying degree of depletion of the cathodes to be treated, which complicates the calculation of precise lithium source requirements, are restricting the large-scale deployment of these processes.

4. Technological comparison among different recycling routes

The majority of mature spent LFP recovery techniques conducted by companies and laboratories rely on pyro/hydrometallurgy and direct restoration. Fig. 4 provides a qualitative comparison among the processes described so far. Direct regeneration, which is the least energy-intensive process, has received considerable attention as a non-destructive, environmentally friendly recycling process [100]. However, it has not yet been applied industrially due to limitations associated with the relative impurity of the recovered cathode and its lack of flexibility in terms of feed materials [101]. Mechanochemical and hydrometallurgical routes, on the other hand, can provide high-quality products with reduced environmental implications provided that non-polluting chemicals are used as leaching and co-grinding agents, respectively. Lastly, pyrometallurgy appears to be the most flexible and scalable technique but is characterized by high energy consumption and low product quality which hinder its large-scale diffusion.

Understanding which, among the presented recycling routes, is the most promising technology is a non-trivial task at the moment as it is difficult to find information on industrial applications relating to the recycling of LFP. The main reason for this difficulty is the fact that the majority of LFP batteries are still in service, and the retirement wave has not yet arrived. Therefore, large-scale industrial recycling is still in its infancy. From an engineering point of view, factors such as cost and duration of production, complexity of the process and equipment and availability of materials should be taken into proper consideration. Attention should be also focused on the environmental impact and damage to human health that could result from the recovery process. Until now, recycling has only been addressed on a laboratory scale and the transition to an industrial scale should be carefully evaluated. It seems logical that recovery processes that involve a high consumption of raw materials and a high degree of complexity are less competitive. Although pyrometallurgical recycling is applicable on an industrial scale due to its simple and mature process, recycling costs are high due to the large energy consumption and complexity of metal separation processes. This becomes more important for LFP cathodes, where the recovery of the low-value metal (iron) does not contribute to the economy of the process. For this reason, direct regeneration represents a solution capable of providing a material with good electrochemical performance through an ecological, simple, and sustainable process. The complexity of the equipment and operations limits this regeneration technique. However, with advances in scientific and technological progress, these methods still have the potential for industrial application.

5. Battery recycling policies

The regulations on the recycling of LIBs in various countries of the world are discussed in some articles that focus mainly on those in force in the United States, the European Union and China due to their relevance on the LIB market [29,30]. It is interesting to note that the presence of adequate regulations for the reuse, disposal and recycling of LIBs could have important environmental and economic impacts.

In the United States, federal, state, and local governments have authority over the disposal and recycling of lithium-ion batteries. Unfortunately, at the federal level, no policy has directly addressed the decommissioning of LIBs, nor does it mandate or incentivize their reuse/recovery [31]. Instead, several bills have been passed since 2020 to fund investments in LIB recycling [32]. Although the law does not mandate battery recycling, several million dollars have been allocated to fund recycling and reuse programs for electric vehicle batteries, and guidelines have been proposed to hold manufacturers accountable, develop voluntary battery labelling guidelines, and develop best practices for

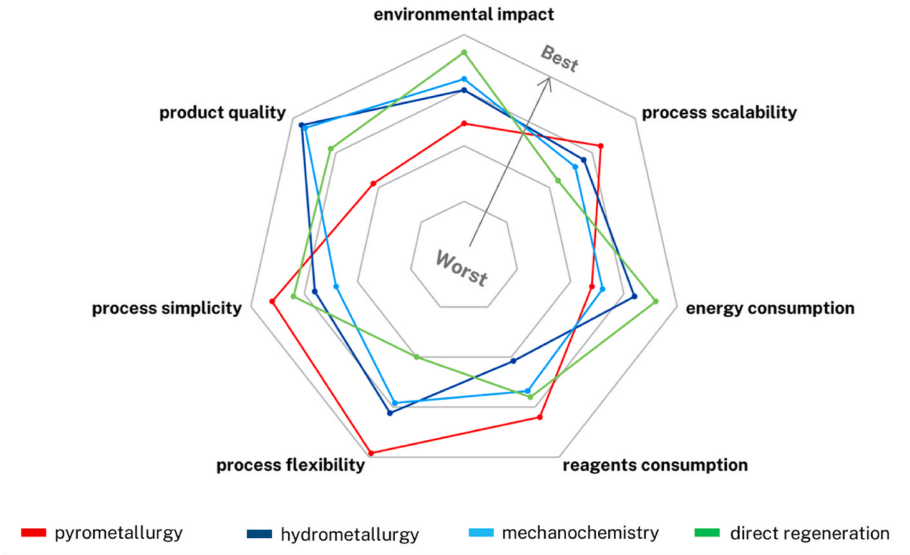

Fig. 4. Comparison of various spent LFP cathode recycling strategies.

battery recycling [33]. In 2006, the European Union enacted the Battery Directive (Directive 2006/66/EC) and the Waste Electrical and Electronic Equipment (WEEE) Directive (Directive 2012/19/EU) requiring member states to set up collection systems for portable batteries in the vicinity of end-users. Subsequently, as part of the European Green Deal, the European Commission presented a legislative proposal in 2020 that significantly surpasses previous legislation in many respects. In this new directive, collection targets for spent portable batteries are raised to 45% by 2023, 65% by 2025 and 70% by 2030. While the EU currently has no regulations governing the recycling of large-scale lithium-ion batteries, it has developed a draft regulation that provides a comprehensive framework for the design, sale, use and recycling of batteries, in particular lithium-ion batteries [34]. The regulation defines the responsibilities of battery producers, users, and recyclers. It also requires "battery passports" that allow end-users to know what a battery contains, where the materials come from and what their environmental impacts are [35] so that their reuse and recycling can be monitored. China has long had a series of regulations and policies to manage solid and hazardous waste, mainly from lead-acid batteries. Later, with the spread of LIBs, regulations began to include these batteries as well. In order to promote the recycling of power storage batteries for new energy vehicles, and to guide and standardize the construction and operation of power storage battery recycling service outlets, the Department of Energy Saving and Comprehensive Utilization of the Ministry of Industry and Information Technology has studied and drafted the Guide to the Construction and Operation of Power Storage Battery Recycling Service sites for New Energy vehicles [36]. The regulations cover many aspects of LIBs production, collection, and recycling. In Japan, there is a non-profit organization (JBRC - Japan Portable Rechargeable Battery Recycling Center) that manages the recycling of Japanese batteries [37]. At the same time, there is also a program to give a second life to batteries (reuse of used batteries for home emergency power). India, despite having regulations for electronic wastes, has not developed any plan for the treatment or recycling of end-of-life LIBs [38]. This could be a serious problem for India, as the lack of a framework for LIBs recycling could make it a potential destination for battery recycling waste. Australia, like India, has not developed any LIB recycling policy [39].

6. Conclusions

This review set out to explore the most relevant strategies for spent LFP batteries recycling, highlighting novelty, limitations and possible improvement opportunities. The main conclusions that can be drawn from this analysis are.

- Proper pre-treatments are crucial to ensure the success of cathode recycling techniques, as they determine the properties of the materials to be processed.
- Pyrometallurgical processes are the most mature ones for the industrial scale but result uneconomical when applied to the relatively cheap LFP cathode.
- Hydrometallurgy and mechanochemistry allow for selective Li recovery, which represents a way to enhance the economic performance of the process. Furthermore, with the use of eco-friendly reactants, they may also be environmentally sustainable.
- Direct regeneration techniques allow for effective cathode restoration with reduced energy and reactants consumption, but their large-scale implementation is currently limited by their reduced flexibility in terms of input materials.
- Due attention should be devoted to the development of specific laws regulating the production and disposal of LIBs, in order to foster a manufacturing uniformity that may facilitate recycling operations and enable the development of large-scale plants.

This review is expected to serve as a reference to navigate the current LFP battery recycling scenario, demonstrating unresolved issues that might improve its sustainable development.

CRediT authorship contribution statement

B. Visone: Writing – review & editing, Writing – original draft, Investigation, Data curation. **O. Senneca:** Writing – review & editing, Conceptualization. **P.P. Prosini:** Writing – review & editing. **B. Apicella:** Writing – review & editing, Supervision, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Nomenclature

Acronyms/abbreviations

CEI	Cathode electrolyte interface
CTR	Carbothermic reduction
EDTA-2Na	Ethylenediaminetetraacetic acid disodium salt
EV	Electric vehicle
FP	Iron phosphate
IEA	International Energy Agency
LCO	Lithium Cobalt oxide
LFP	Lithium Iron phosphate
LIB	Lithium-ion battery
Li_2DHBN	3,4-dihydroxybenzonitrile dilithium
LMO	Lithium Manganese oxide
NCM	Nickel Manganese Cobalt oxide
PVDF	Polyvinylidene fluoride
SEI	Solid electrolyte interface

Data availability

Data will be made available on request.

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