

Precision Recycling of Spent Cathode Materials: From Degradation Mechanisms to Recycling Strategies

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Abstract: The increasing number of spent lithium-ion batteries has raised growing concerns regarding resource supply and environmental impacts, making the efficient recycling of high-value cathode materials an important issue in battery research. This review examines the relationship between material degradation and recycling strategy, highlighting that the recycling route should be selected according to the actual failure features of the cathode. Major degradation modes, including lithium loss, surface/interface deterioration, transition-metal dissolution, structural disorder, and phase reconstruction, are first summarized, followed by discussion of their effects on different recycling approaches. On the basis of these analyses, a mechanism-guided framework for strategy selection is proposed. The review also briefly discusses the possible role of artificial intelligence and digital twin technologies in improving failure diagnosis, process design, and recycling optimization, with the aim of promoting efficient and sustainable closed-loop recycling of spent cathodes.

Key words: lithium-ion batteries; cathode materials recycling; degradation mechanisms; recycling strategy; closed-loop recycling

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1. Introduction

The global energy transition has promoted the unprecedented deployment of lithium-ion batteries (LIBs). According to the sustainability scenarios described in the Shared Socioeconomic Pathways (SSP1), the demand for LIBs is expected to reach 10,000 GWh in 2040^[1,2]. However, such large-scale deployment will inevitably lead to a sharp increase in decommissioned batteries. For electric vehicle LIBs alone, up to 8 million tons of waste are estimated to be generated by 2040^[3]. Under China's carbon neutrality (CN60) goal, 46 million tons (46 Mt) of spent LIBs are projected by 2060^[4], posing serious resource and environmental challenges (Figure 1a). Among battery components, the cathode materials act as the main carriers of cost and value^[5], containing high amounts of critical metals. Meanwhile, in response to the demand for high capacity, layered oxide cathode materials have become the dominant component^[6] (Figure 1b). Therefore, the efficient recycling of spent cathode materials is not only necessary for environmental protection and sustainable development, but also important for easing the supply pressure of strategic metal resources (such

as lithium, cobalt, nickel, and manganese)^[7,8]. Their effective recycling is therefore highly important for achieving resource circulation and improving economic value. Furthermore, predictions based on the lithium cycle computable general equilibrium (LCCGE) model indicate that by 2060, consumption-oriented recycling will reduce the global emission intensity by 16.30%^[9] (Figure 1c).

Ideal recycling protocols for cathode materials aim to maximize the recovery and circular use of valuable materials while reducing energy consumption and environmental impacts. At present, the main recycling technologies can be divided into five types: pyrometallurgical, hydrometallurgical, direct recycling/upcycling and emerging strategy^[10–12]. These methods show clear significant carbon-reduction advantages, decreasing greenhouse gas emissions across the full battery lifecycle by about 17%, 51%, and 61%^[13], respectively, and thus effectively reducing the carbon footprint of the whole supply chain.

Although recycling technologies have made considerable progress, current recycling strategies still often depend on rough sorting and processing approaches^[14,15]. As a consequence, this not only affects the efficient recovery of high-value materials but also restricts the technical and economic feasibility of a closed-loop material system. Therefore, increasing attention has been given to the accurate identification and targeted matching of recycling methods for cathode materials under real failure conditions^[6,16,17]. While several recent high-quality reviews have provided valuable insights into lithium-ion battery recycling, they predominantly focus on general

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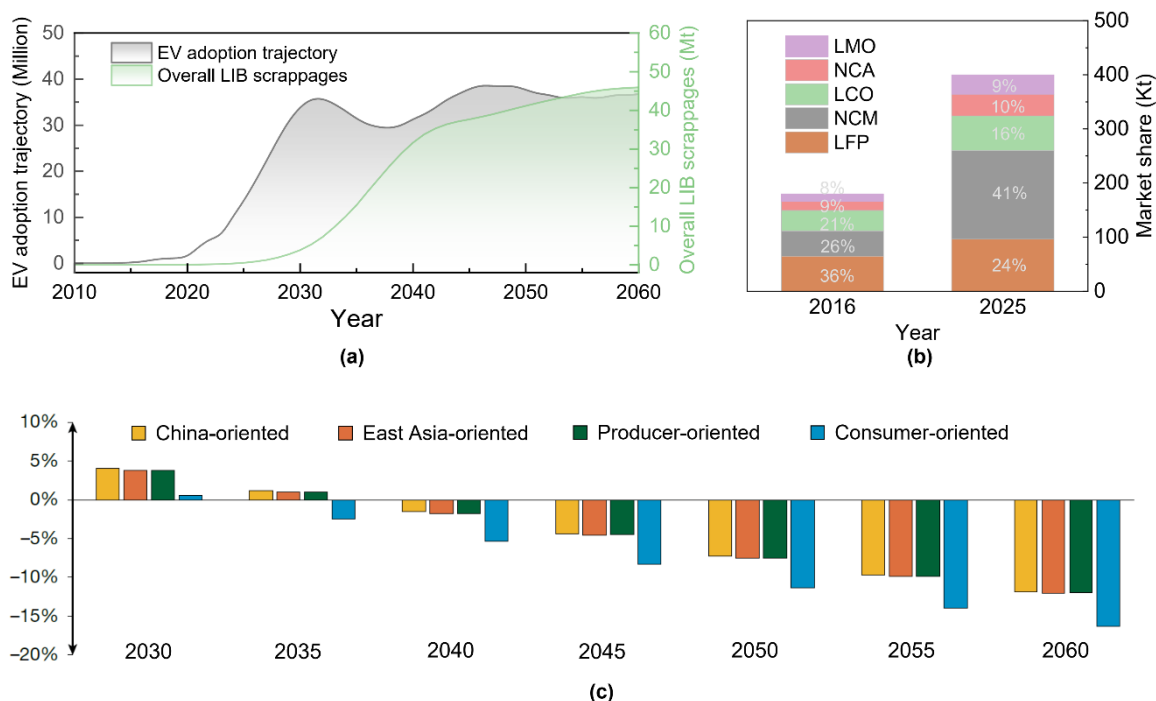


Fig. 1. Market and environmental landscape of LIBs. (a) Electric vehicle (EV) adoption trends from 2010 to 2060 and overall LIBs scrappages; reprinted from Ref.^[4]. (b) Market shares of five typical commercial cathodes in 2016 and 2025; reprinted from Ref.^[6]. (c) The rate of change in global greenhouse gas emission intensity during the period from 2030 to 2060; reprinted from Ref.^[9].

material degradation phenomena, technological progress, and sustainability analyses. For example, previous works have systematically discussed failure mechanisms across different scales^[6], detailed mainstream recycling technologies and their environmental impacts^[11,14], and comprehensively evaluated the cascading utilization of retired batteries^[16]. However, comprehensive discussions explicitly linking specific cathode degradation states with the most suitable recycling strategies remain remarkably scarce. To fill this critical gap, this review introduces a novel “mechanism-to-strategy matching” framework.

In this review, the focus is placed on the “matching of material failure mechanisms and recycling strategies.” First, the typical failure mechanisms of lithium-ion batteries are systematically examined, particularly for cathode materials, including layered oxides (such as NCM and LCO), olivine-type materials (lithium iron phosphate), and spinel-type materials (lithium manganese oxide). Subsequently, a comprehensive overview is provided of the current major battery recycling strategies, together with their basic principles, advantages, and disadvantages. On this basis, how the specific failure states of cathode materials can be considered together with the characteristics of different recycling strategies to realize optimized selection and targeted matching of strategies is further discussed. Next, the compatibility between cathode materials with different failure characteristics and various recycling schemes is analyzed through case studies. Finally, with attention to future technological frontiers, the application of advanced multimodal artificial intelligence in battery recycling is discussed, and how artificial intelligence and digital twin technology may support intelligent diagnosis of failure states, precise design of recycling processes, and dynamic optimization is proposed, ultimately promoting the establishment of a closed-loop system for “non-

destructive/micro-destructive failure analysis - intelligent decision-making - precise recycling.”

2. Material degradation mechanisms

The degradation of lithium-ion batteries (LIBs) is a complex and irreversible process driven by coupled electrochemical, thermal, and mechanical stresses, resulting in performance decay from atomic to macroscopic scales^[17–19]. Understanding these failure mechanisms is essential for developing efficient, precise, and green recycling strategies^[6,10,16]. As illustrated in **Figure 2a**, multiple of physical and chemical mechanisms jointly contribute to cell failure, affecting all components, including electrodes, electrolyte, separator, and current collectors. Common degradation mechanisms can be systematically classified into structural degradation (e.g., particle cracking, irreversible phase transitions), chemical/compositional degradation (e.g., transition metal (TM) dissolution, oxygen release, and lithium inventory loss), and interfacial degradation (e.g., CEI (cathode electrolyte interface) thickening, electrode/electrolyte side reactions)^[19]. The diversity, interdependence, and different evolution rates of these mechanisms make degradation modeling highly challenging. Therefore, a detailed understanding of LIB degradation mechanisms is important for enabling safe and efficient recovery and selective resource reclamation.

The degradation behavior of LIBs is closely associated with the properties of cathode material. **Figure 2b** shows typical intercalation-type cathode materials widely used in commercial power batteries, together with their crystal structures. Currently, the main cathode materials include: layered metal oxides (e.g., LiCoO_2 -LCO, $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ -NCM, lithium-rich manganese-based layered oxides-LMR)^[20–24], olivine-type materials (LiFePO_4 -LFP)^[25–27], and spinel-type materials (LiMn_2O_4 -

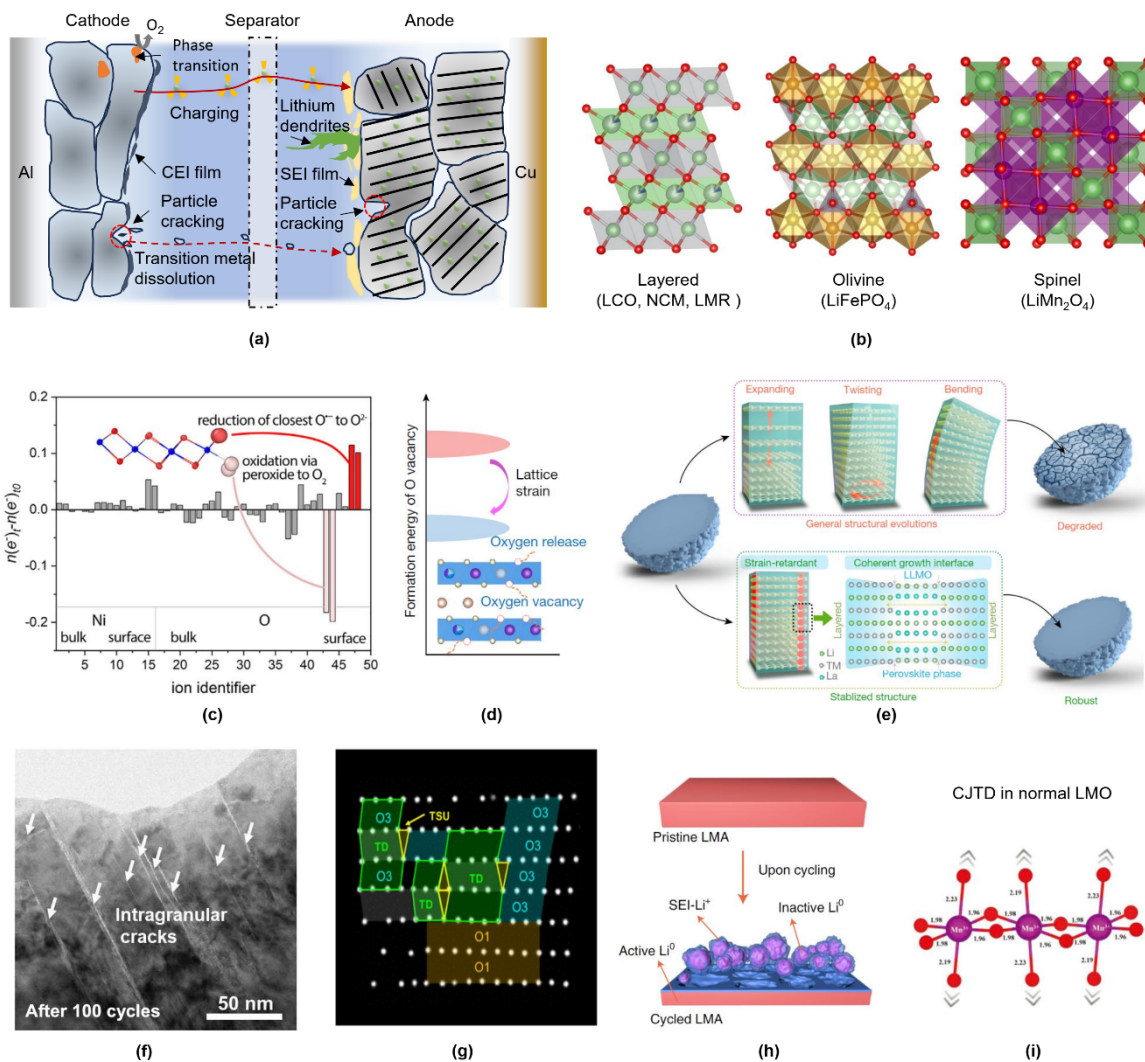


Fig. 2. Degradation mechanisms. (a) Schematic illustration of the degradation mechanisms of LIBs, such as cathode phase transitions, transition metal dissolution, and particle cracking. (b) Schematic diagram of the crystal structures of three typical oxide cathode materials (layered, olivine, and spinel); adapted from Ref. [28]. (c) Mechanistic analysis of the observed route of O_2 evolution and the formation of $[1]O_2$; adapted from Ref. [40]. (d) Schematic illustrating how lattice strain fundamentally lowers the formation energy of oxygen vacancies, thereby accelerating oxygen release; adapted from Ref. [41]. (e) Illustration of lattice distortion (expanding, twisting, bending) leading to severe macroscopic particle degradation; adapted from Ref. [47]. (f) TEM images of SC-NMC after 100 cycles, showing intragranular cracks; adapted from Ref. [49]. (g) ADF-STEM image of a delithiated NMC-811 cathode showing the formation of continuous (C-type) and abrupt (A-type) O1–O3 interfaces as well as TDs; adapted from Ref. [50]. (h) Schematic of lithium inventory loss, in which the porous irreversible Li layer the containing residual active Li^0 is composed of SEI-encapsulated inactive Li^0 ; adapted from Ref. [53]. (i) Mn^{3+} octahedrons in normal LMO distort along the z axis and generate large structure changes because of the cooperative Jahn–Teller effect; adapted from Ref. [34].

LMO)[28–30]. Throughout their lifecycle, these cathode materials undergo complex and structurally dependent failure mechanisms[19]. For layered oxide cathodes, the degradation is dominated by severe structural degradation —specifically, anisotropic lattice expansion and contraction (i.e., a -axis contraction and c -axis expansion) during cycling, which induces severe microcracking between primary particles. Concurrently, they suffer from profound interfacial degradation, where highly oxidative species (e.g., Ni^{4+}) catalyze continuous electrolyte decomposition, forming a thick and resistive cathode electrolyte interphase (CEI)[31,32]. In contrast, for olivine-type LFP cathodes, the dominant failure mode is chemical/compositional degradation, which predominantly manifests as an irreversible loss of active lithium inventory rather than bulk structural collapse[33]. Finally, for spinel-type LMO cathodes, the pri-

mary mechanism involves severe chemical/compositional degradation closely coupled with structural distortion, primarily driven by manganese dissolution initiated by the Jahn–Teller effect[34].

As shown in **Figures 2c–i**, the degradation of cathode materials occurs through several atomic- and particle-scale mechanisms. Various theories have been proposed to explain the failure mechanisms of cathode materials caused by oxygen release[35–39]. Using computational tools such as density functional theory (DFT) and dynamical mean field theory (DMFT), Morris et al.[40] successfully captured singlet oxygen formation on the cathode surface and analyzed the spontaneous release of oxygen (O_2) from the (012) surface of delithiated nickel-rich cathode materials (**Figure 2c**). LMR layered cathode materials, such as $Li_{1.2}Ni_{0.13}Mn_{0.54}Co_{0.13}O_2$, have received considerable

attention owing to their high energy density. Amine et al.^[41] demonstrated that the evolution of lattice strain leads to structural degradation and oxygen loss, thereby inducing voltage decay (Figure 2d). During charge and discharge processes, lithium (de)intercalation in layered oxide cathode materials causes anisotropic lattice strain, leading to intragranular crack formation and particle fracture^[42] (Figure 2e and 2f). This degradation becomes more serious under high-voltage conditions^[43–46]. Lu et al.^[47] introduced a coherent perovskite phase that acts as “rivets,” thereby stabilizing the structure. Amine et al.^[48,49] systematically investigated the relationship between stress evolution and failure mechanisms in both single-crystal and polycrystalline layered nickel-rich cathode materials. As illustrated in Figure 2g, Xin et al.^[50], using artificial intelligence-enhanced super-resolution electron microscopy, found that the coupling between confinement and phase transitions induced by interlayer shear aggravates the degradation and failure of layered cathodes. During delithiation, a complex O1-O3 mixed phase forms on the surface of cathode particles (NMC811). Furthermore, Lu et al.^[47] tracked the H1-M-H2-H3 phase transition in P-NCM by combining in-operando synchrotron XRD with ex-situ high-resolution XRD, revealing that this phase transition is accompanied by significant lattice strain. Distinguishing reversible lithium from irreversible lithium is essential for assessing the reversibility of lithium metal batteries^[51,52]. Liu et al.^[53] quantitatively evaluated reversibility in practical lithium metal batteries by determining the mass of active and inactive lithium after cycling (Figure 2h). In LMO cathode particles, Jahn-Teller distortion (Figure 2i) promotes manganese dissolution^[34,54,55] and irreversible phase transformation (from cubic to tetragonal phase)^[56–58]. Based on an in-depth analysis of the failure mechanisms of cathode materials, targeted recycling strategies can be designed for spent materials with specific aging characteristics.

3. Recycling strategies for spent cathodes

Taking a typical pouch cell as an example (Figure 3a), the

basic structure of a LIB mainly consists of metal casing components, electrode active materials, a separator, and electrolyte, all of which have recyclable value and require disassembly before recycling^[59–61]. The casing components include the tab, laminated film, current collectors (copper foil for the anode and aluminum foil for the cathode), and the battery can^[62]. Regarding electrode active materials, the cathode mainly consists of lithium-containing transition metal oxides with high recycling value, such as LCO, NCM^[63,64], LMO, or LFP^[25,28,62,65]. The anode is generally composed of graphite^[66–68] or lithium metal^[69–71]. The separator is a porous polymer membrane (e.g., polyethylene, polypropylene) positioned between the cathode and anode^[72,73]. The electrolyte is prepared by dissolving a lithium salt^[74,75] (e.g., lithium hexafluorophosphate, LiPF_6) in organic solvents (e.g., a mixture of ethylene carbonate and dimethyl carbonate), serving as the medium for ion conduction^[76,77]. Therefore, the central objective of the recycling process is to safely, efficiently, and economically separate and regenerate high-value materials from these complex components.

At present, according to the treatment methods used for spent battery materials and the corresponding target products, recycling technologies are mainly classified into five major categories: pyrometallurgy, hydrometallurgy, direct recycling, upcycling and non-destructive direct regeneration. A detailed comparison of different recycling strategies is shown in Table 1. As shown in Figure 3b, pyrometallurgical recycling is a high-temperature treatment process usually carried out at temperatures above 1000°C^[78,79]. This method involves feeding spent batteries into high-temperature furnaces such as electric arc furnaces or rotary kilns for smelting, commonly in the presence of reducing agents such as coke. During this process, organic components such as electrolytes, binders, and separators are thermally decomposed and combusted, while metal oxides are reduced. This finally produces a molten alloy mainly composed of cobalt, nickel, copper, etc., and slag enriched with elements such as lithium, aluminum, manganese, and phosphorus^[80,81]. The alloy can be further refined

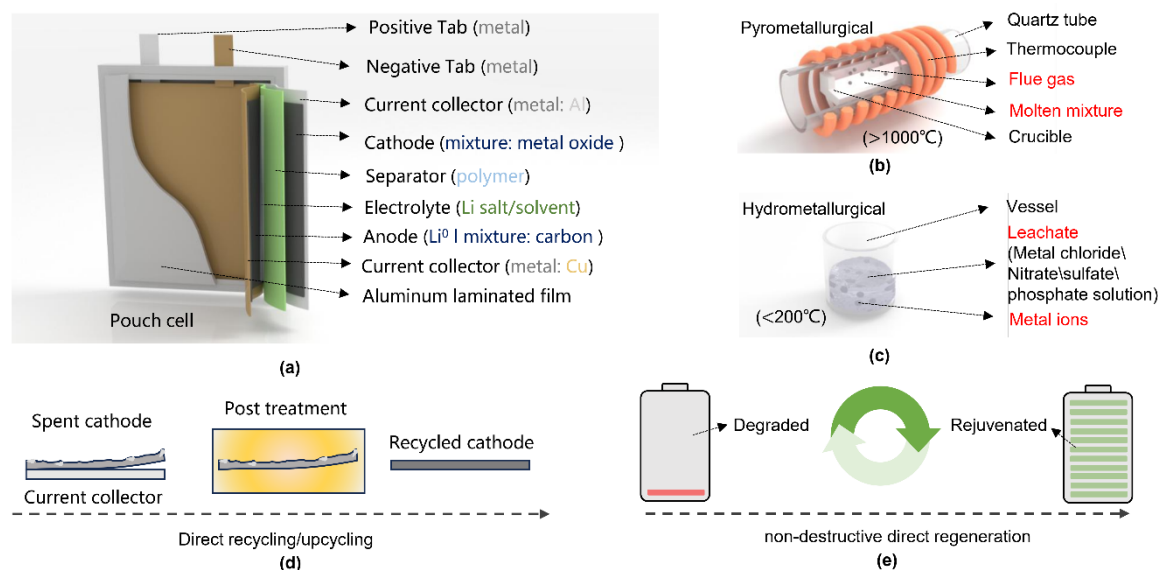


Fig. 3. Recycling schemes. (a) Components and structure of a pouch cell. (b–e) Different recycling strategies: (b) pyrometallurgy, (c) hydrometallurgy, (d) direct recycling/upcycling, and (e) non-destructive direct regeneration.

Table 1. Systematic comparison of different recycling strategies^[16,79,104–109]

Indicator	Pyrometallurgy	Hydrometallurgy	Direct recycling	Upcycling	Non-destructive direct regeneration
Metal recovery rates	Co, Ni: >95% (to alloy); Li: <10% (to slag); Mn: 30–50% (to slag)	Li: 85–95%; Co, Ni: 90–98%; Mn: 85–95% (all as battery-grade salts)	Li: 90–98% (supplemented); Co, Ni, Mn: >97% (retained in structure)	Li: 85–95%; TM: >95% (reincorporated into upgraded product)	Li: 98–100% (in-situ replenished) All other elements 100% retained in intact electrode structure
Energy consumption (kWh/kg cathode)	~15–30	~8–20	~2–10	~5–12	~0.5–2
GHG emissions (kg CO ₂ -eq/kg cathode)	~1–3	~0.5–2	~1–3	~1–3	~0.3–1.5
Operational complexity	Low–moderate	Moderate–high	Moderate–high	High	Low–moderate
Industrial maturity (TRL)	Commercial (TRL 8–9)	Commercial (TRL 8–9)	Lab to pilot (TRL 4–6)	Lab to pilot (TRL 3–5)	Lab scale (TRL 1–3)
Economic feasibility (\$/kg net)	–4 to +0.5	–2 to +1.5	+3 to +8	+5 to +12	+8 to +15

Note: Regarding metal recovery rates, the values for non-destructive direct regeneration, direct recycling, and upcycling refer to the fraction of metal retained in the regenerated cathode material relative to the spent input, rather than elemental extraction yields. For pyrometallurgy and hydrometallurgy, the values represent the final yield of separated metals or metal salt products.

and purified. This technology has advantages such as a relatively simple process flow and low requirements for feedstock pretreatment; it can directly process whole batteries without prior discharging and dismantling. However, it also has limitations, including high energy consumption and the production of hazardous exhaust gases, requiring complicated exhaust gas treatment systems^[82,83].

As illustrated in **Figure 3c**, hydrometallurgical recycling is a solution-based chemical processing method performed under mild conditions, generally below 200°C^[78,84]. This method starts with leaching the preprocessed cathode black mass using acids (e.g., sulfuric acid, hydrochloric acid) or alkaline solutions, thereby dissolving valuable metals into ionic forms to produce a leachate containing metal chlorides, nitrates, sulfates, or phosphates^[85–87]. Subsequently, through a series of chemical separation steps such as solvent extraction^[88,89], precipitation, crystallization, and electrodeposition^[90–92], target metal ions such as nickel, cobalt, manganese, and lithium are separately recovered and purified from the complex leachate. Finally, high-purity metal salts or precursors are obtained, which can be directly used to resynthesize cathode materials. The advantages of hydrometallurgy include relatively high metal recovery rates, especially for lithium, and high product purity, making it a mainstream technology in both current research and industrial applications. However, this method also has disadvantages, including long processing steps, high consumption of chemical reagents, and the generation of large amounts of waste leachate and residues that require further treatment^[93–95].

The core of direct recycling and upcycling lies in repairing the structural defects of degraded cathode materials (**Figure 3d**), rather than breaking them down into their constituent elements or simple compounds. This technology directly restores the stoichiometry, crystal structure, and electrochemical performance of preliminarily separated and purified spent cathode materials through a series of post-treatments such as solid-state sintering^[96,97], hydrothermal methods^[98,99], eutectic salt methods^[100], or electrochemical methods^[10,101]. At the same time, stabilizing the lattice structure of the recovered cath-

odes remains critical for direct recycling strategies to yield materials with prolonged operational lifespans^[102,103]. Furthermore, the non-destructive direct regeneration shown in **Figure 3e** represents a more “in situ” repair strategy. It aims to recover the performance of degraded batteries without damaging their original device structure. This method is a highly promising sustainable route, as it avoids the need for complex disassembly, separation, and remanufacturing steps^[10]. However, this technology is still at the laboratory exploration stage. It places extremely high requirements on the understanding of material failure mechanisms, and the stability of its repair efficacy effect as well as the general applicability of the process still need further validation.

4. Selection of recycling strategies

As illustrated in the flowchart of **Figure 4**, this review provides a comprehensive framework for the systematic evaluation and selection of recycling strategies for spent lithium-ion batteries. With the failure state of cathode materials used as the core criterion, this framework aims to guide the selection of the most efficient and cost-effective recycling pathway. The main process begins with spent lithium-ion batteries, which first undergo a standardized pretreatment stage. This pretreatment usually includes the following steps: electrochemical performance evaluation (to preliminarily assess the overall health status and remaining value of the battery)^[110,111], safe discharging (to remove residual charge and ensure the safety of subsequent operations)^[112–114], and disassembly and sorting (where battery components such as the casing, electrodes, and separator are dismantled, with fine sorting specifically applied to cathode materials of different chemical compositions)^[115,116]. Crucially, during these dismantling and pretreatment steps, various impurity elements are inevitably introduced into the recycling stream. These impurities are conventionally considered detrimental; if not effectively removed during direct recycling, they will continuously accumulate over multiple recycling cycles, thereby severely impairing the electrochemical performance of the regenerated materials^[117]. However, some impurities, such as Al³⁺, Cu²⁺, and F[–], can actually enhance the perfor-

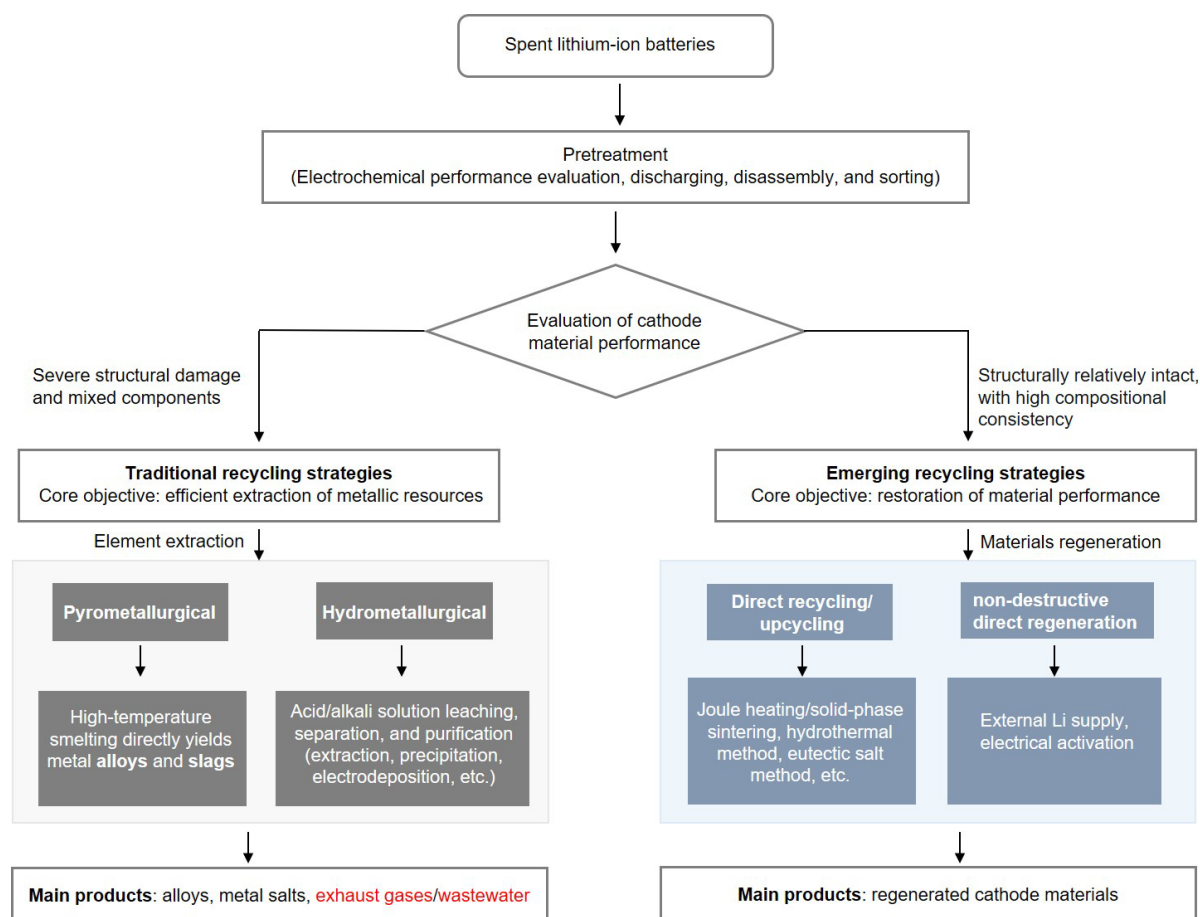


Fig. 4. Comprehensive evaluation of recycling strategies.

mance of cathode materials when present at appropriate concentrations and in specific chemical states^[118].

After pretreatment, the core decision point of the framework is the evaluation of the performance of the separated cathode materials. Furthermore, we introduce a quantifiable evaluation framework. As proposed in Table 2, we establish a six-factor quantitative scoring system (on a scale of 1 to 5) that integrates intrinsic material degradation metrics (i.e., degree of lithium loss, structural collapse severity, and transition-metal dissolution extent) with extrinsic techno-economic parameters (i.e., recovery efficiency, energy consumption, and economic value). Building upon these quantitative indicators, Table 3 presents a comprehensive decision framework that systematically translates complex degradation fingerprints into standardized numerical scores. Through detailed characterization of

their physicochemical properties - such as structural integrity, compositional consistency, and failure mechanisms - and guided by this scoring matrix, the materials are then directed toward two fundamentally different recycling routes.

Route 1, termed traditional recycling, is designed for cathode materials that have undergone severe structural damage and contain mixed compositions - typically corresponding to material health scores of ≤ 2 in Table 3 - such as those experiencing significant phase transformations, particle pulverization, or extensive multi-metal dissolution^[11,14]. Its primary goal is to efficiently extract metal resources rather than restore the original structure of the materials. The main methods used include pyrometallurgy, which produces metal alloys and slag through high-temperature smelting but suffers from high energy consumption and lithium loss (Figure 3b), and

Table 2. Quantitative scoring framework for the degradation states and recycling viability of spent cathodes

Score	Lithium loss degree (Li deficiency)	Structural collapse severity (I(003)/I(104) or rock-salt phase)	TM dissolution extent (in electrolyte or on anode)	Recovery efficiency (target product yield)	Energy consumption (kWh/kg product)	Economic value (net profit, \$/kg feedstock)
5	<5% deficiency	I(003)/I(104) > 1.3, no rock-salt phase	<200 ppm	>95%	<8	> +8
4	5–10% deficiency	1.0–1.3, trace disorder	200–500 ppm	90–95%	8–15	+3 to +8
3	10–20% deficiency	0.7–1.0, localized rock-salt phase	500–1000 ppm	80–90%	15–30	–2 to +3
2	20–35% deficiency	0.4–0.7, significant rock-salt phase	1000–3000 ppm	65–80%	30–50	–5 to –2
1	>35% deficiency or unidentifiable	<0.4 or multiple impurity phases	>3000 ppm	<65%	>50	< –5

Table 3. Decision framework for matching recycling strategies with specific cathode failure states.

Recycling strategies	Li loss tolerance	Structural requirement	TM dissolution tolerance	Recovery efficiency	Energy consumption	Economic value	Recommended weighted total score range
non-destructive direct regeneration	5	5	4	5	5	5	4.5-5.0
Direct recycling	4	4	3	5	4	4	3.5-4.5
Upcycling	3	3	3	4	3	5	3.0-4.0
Hydrometallurgy	2	2	2	4	2	3	2.0-3.0
Pyrometallurgy	1	1	3	2	1	2	1.0-2.0

hydrometallurgy, which uses acid or alkali leaching followed by separation and purification processes to extract metals in ionic form (Figure 3c). Although these methods ultimately yield alloys and metal salts, they also produce exhaust gases and wastewater that require proper treatment. The final products can serve as essential chemical raw materials for the resynthesis of cathode precursors or for other industrial applications^[119–121].

Route 2, known as emerging recycling, is suitable for cathode materials that retain a relatively intact structure and show high compositional consistency - typically requiring material health scores of ≥ 3 - such as homogeneous materials that have undergone only mild lithium loss and surface degradation while maintaining their bulk crystal lattice^[11,14]. The main objective of this route is to restore the performance of these materials, allowing their direct reuse in the production of new batteries. The primary methods used include direct recycling, or upcycling, which replenishes lithium and repairs degraded cathode materials using techniques such as Joule heating, solid-phase sintering, hydrothermal treatment, and eutectic

salt methods to restore the structural integrity and stoichiometry of the material (Figure 3d). Another approach, known as non-destructive direct regeneration, aims to recover performance at the device level while maintaining the original electrode structure, using methods such as supplying lithium from an external source or employing electrochemical activation (Figure 3e). The final products of these processes are regenerated cathode materials that can either be used directly or undergo minimal processing for the manufacture of new batteries^[101,122].

5. Recycling of typical cathode materials

The optimal selection of a recycling strategy largely depends on the failure state and inherent chemical characteristics of the cathode material. Pyrometallurgy uniformly smelts various complex mixed materials, resulting in low specificity toward different failure states^[79]. Therefore, it is not the primary focus of this chapter. The following discussion concentrates on hydrometallurgy, direct recycling, upcycling and non-destructive direct regeneration strategies, which provide

Table 4. Recycling strategies corresponding to different battery health states

Materials	State of health	Main reagents	Experimental conditions	Regeneration methods	Refs.
LiNi _{0.5} Co _{0.2} Mn _{0.3} O ₂ (NCM523)	80% or less of the rated capacity	LiOH	Solid-state sintering treatment	Direct recycling	[123]
LiCoO ₂	Discarded cell phone battery	MgO, Al ₂ O ₃	Heated to 900°C in air	Upcycling	[105]
LiCoO ₂		Lithium lignosulfonate	Heated to 850°C for 6 h	Upcycling	[126]
LiCoO ₂		LiCoO ₂ , NiC ₄ H ₆ O ₄ ·4H ₂ O, MnC ₄ H ₆ O ₄ ·4H ₂ O, LiOH·H ₂ O	Annealed at 850°C and second sintering at 900°C	Upcycling	[127]
LiNi _{1-x-y} Mn _x Co _y O ₂ (NMC)	Spent LIBs with an NMC cathode	NMP, oxalic acid	heated to 95°C	Hydrometallurgical (acid leaching and radiative heating)	[130]
NCM622, NCM523, NCM111 and LiCoO ₂ (LCO)	Spent materials purchased from Zhejiang Huayou Cobalt Co., Ltd.	H ₂ O, glycine, NaOH	heated to 80°C in a water bath	Hydrometallurgical	[134]
LiFePO ₄	Degraded to less than 80% of the initial value	Li ₂ C ₂ O ₄ , CMK-3	Prelithiation separator	Direct recycling	[138]
LiFePO ₄		LiSO ₂ CF ₃	External lithium supply approach	Non-destructive direct regeneration	[104]
LiFePO ₄	LG (IFR18650), 3.2 V, 1.2 Ah SP-LFP and SP-Gr obtained from spent batteries supplied by Yingwang Battery; batteries were discharged to 1 V before disassembly	Li ₂ DHBN	Heated to 800°C	Direct recycling	[107]
LiFePO ₄		Li ₂ CO ₃ , glucose	Heated at 700°C	Direct recycling	[106]
LiMn ₂ O ₄ \ LiNi _{0.5} Mn _{1.5} O ₄	Spent lithium manganese pouch cell (LiMn ₂ O ₄ , Ronbay)	LiOH, Li ₂ CO ₃	Heated to 850°C	Upcycling	[150]
LiMn ₂ O ₄	Spent LMO purchased from Quzhou Huayou Cobalt New Material Co., Ltd.	Na ₂ SO ₃ , H ₂ O, Na ₂ CO ₃	Water bath preheated to 90°C	Hydrometallurgical (oxidation leaching)	[151]
LiMn ₂ O ₄	LIBs tested at 1 C (4 A) for 1000 cycles with capacity retention below 80%, regarded as spent LIBs	H ₂ SO ₄ , H ₂ O ₂	Acid leaching	Hydrometallurgical	[154]

more precise control according to the failure state of the material. **Table 4** systematically summarizes the recommended recycling approaches corresponding to different failure states.

5.1. Recycling of layered cathode materials

For degraded layered cathode materials (such as NCM, LCO, etc.) with a relatively intact main structure, the core of direct recycling is to repair structural defects^[123,124], eliminate cation disorder, and reconstruct the ordered layered structure through lithium replenishment and thermal treatment (**Figure 5a**). For example, Liu et al.^[125] proposed a non-destructive regeneration technique based on solid-state sintering, which can effectively restore the crystal structure of degraded LCO. Fang et al.^[126] developed a one-step regeneration strategy using lithium lignosulfonate (Li-L) to efficiently replenish

lithium and restore the structural integrity of LCO. The regenerated LCO showed stable electrochemical performance even under a high voltage of 4.6 V.

Direct recycling technology also provides the advantage of flexibly adjusting the chemical composition of the final product, thereby enabling “upcycling”. For example, Wang et al.^[105] demonstrated a single-step process in which spent LCO directly reacts with aluminum oxide, magnesium oxide, and lithium carbonate to synthesize a more structurally stable LCO cathode material that can operate at 4.6 V. The in-situ XRD contour map in **Figure 5b** shows the gradual intercalation of lithium ions and the restoration of the lattice structure during the sintering process. Yang et al.^[127] designed a mechanochemical strategy to directly upcycle degraded LCO cathode material into NCM cathode material. Overall, these methods can

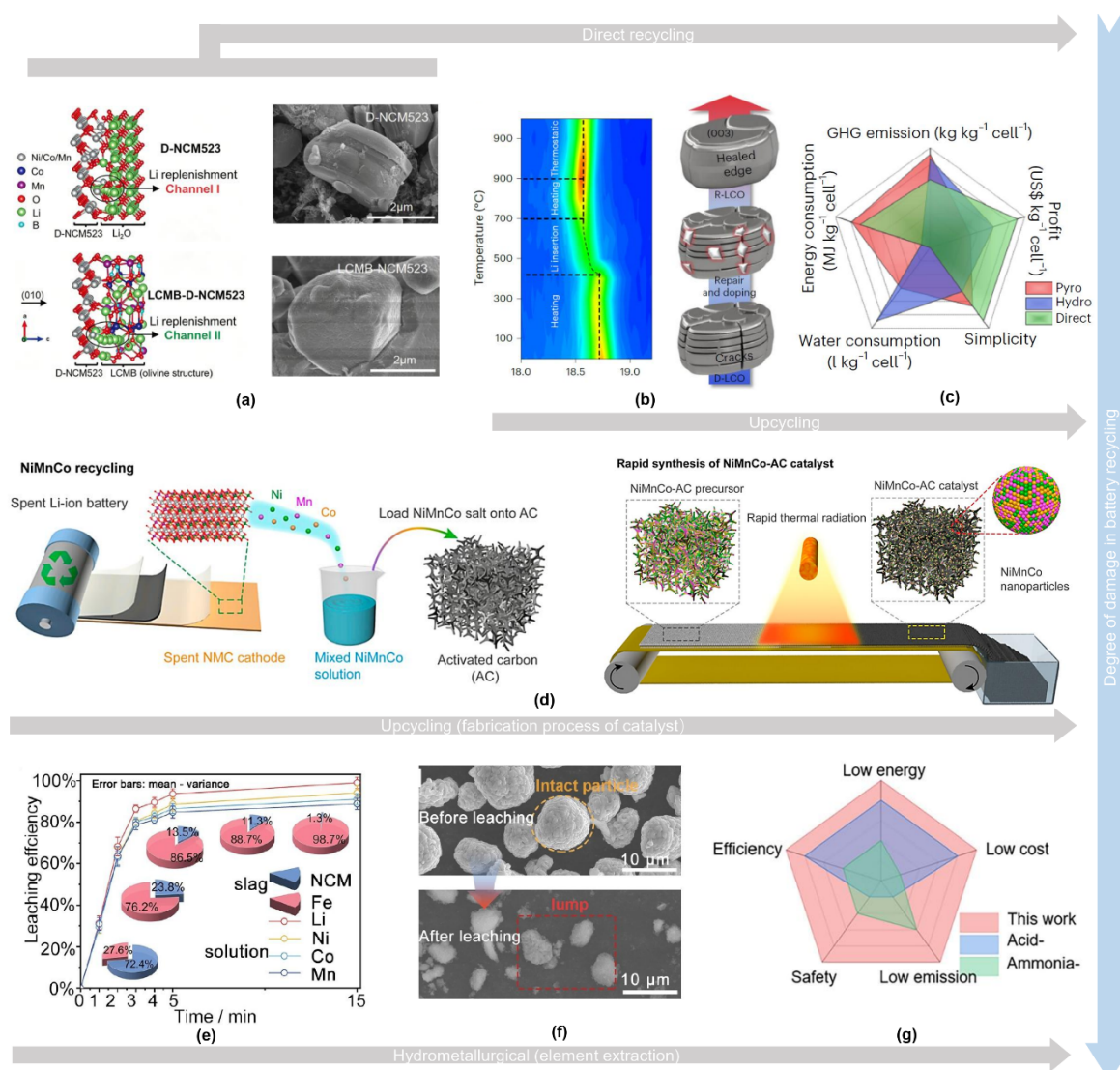


Fig. 5. Recycling of layered cathode. (a) Fast Li replenishment channels enhancing direct recycling technology: 3D models illustrating distinct Li replenishment channels in degraded NCM523 (D-NCM523) and LCMB coating treated D-NCM523 during regeneration, alongside the SEM images of D-NCM523 and LCMB-NCM523; adapted from Ref.^[135]. (b) In situ XRD patterns of degraded LCO during thermal treatment and schematic of the morphology changes of degraded LCO during regeneration, showing the structural healing process; (c) Comprehensive technoeconomic radar chart highlighting the overall superiority of direct recycling over traditional methods in profitability and simplicity; adapted from Ref.^[105]. (d) Schematic of upcycling a value-added NiMnCo-AC catalyst from spent NMC cathodes via rapid thermal radiation; adapted from Ref.^[130]. (e) Hydrometallurgical leaching curves and changes in the composition of the leach slag; (f) Scanning electron microscope images before and after leaching; (g) Comprehensive evaluation radar chart demonstrating the efficiency and environmental metrics of different leaching methods; adapted from Ref.^[134].

markedly reduce greenhouse gas and other waste emissions while improving the economic benefits of the recycling process (Figure 5c).

For severely degraded layered materials that are difficult to repair directly, they can be repurposed as precursors and converted into high-value-added transition metal-based catalysts through chemical treatments^[128,129]. For example, Jiao et al.^[130] used a rapid thermal radiation method, involving acid leaching and radiant heating processes (Figure 5d), to convert spent NMC cathode material into highly efficient nickel-manganese-cobalt-activated carbon (NiMnCo-AC) catalysts for zinc-air batteries. Similarly, Yao et al.^[131] converted CNM811 and LFP materials into electrocatalysts for the oxygen evolution reaction (OER) through leaching and co-precipitation.

Hydrometallurgical extraction of valuable elements is the mainstream industrial approach for treating mixed or severely degraded layered materials. Although conventional hydrometallurgy commonly uses acid^[132] or ammonia^[133] leaching, Xu et al.^[134] proposed a neutral leaching technique utilizing glycine as a coordination agent. This method achieved nearly 100% leaching efficiency for metals from NCM111 (Figure 5e), with the appearance of the material before and after leaching shown in Figure 5f. Compared with acid and ammonia leaching, this neutral leaching strategy shows advantages in energy efficiency, emissions, safety, and profitability (Figure 5g).

5.2. Recycling of LFP cathode materials

LFP has excellent structural stability, and its main degradation mechanism is the loss of active lithium^[109,136]. Non-destructive direct regeneration and direct recycling strategies can efficiently and economically replenish lithium sources^[137]. For example, Chen et al.^[104] developed a method that restores spent batteries without disassembly by externally supplying lithium (Figure 6a). The use of LiSO_2CF_3 as the lithiation material, which is fully converted during cycling, avoids gas evolution (Figure 6b). The cost of battery repair using this method is reduced by two orders of magnitude compared with manufacturing new batteries (Figure 6c). Fan et al.^[138] proposed a functionalized pre-lithiation separator technology that enables in situ electrochemical regeneration of LFP (Figure 6d), effectively recovering its electrochemical performance (Figure 6e). Ji et al.^[107] performed lithium replenishment through solid-state sintering while simultaneously forming a surface carbon layer that improves lithium-ion and electron transport kinetics (Figure 6f). Furthermore, considering that the diffusion kinetics of lithium ions within the degraded LFP lattice are crucial during direct recycling, Xiao et al.^[139] employed polyacrylonitrile (PAN) as a functional additive. This approach elegantly promoted intimate contact between the external lithium source and the degraded LFP, thereby enabling the effective repair of unevenly distributed Fe-Li defects within the degraded material. Direct recycling shows clear advantages over hydrometallurgy and pyrometallurgy in environmental protection and energy conservation (Figure 6h). However, the efficient and green separation of active materials from current collectors during recycling remains difficult. Yang et al.^[106] introduced a water electrolysis-induced separation method (Figure 6g), which is suitable for various electrode types and reduces energy consumption as well as the use of additional acid/alkaline solutions. Figure 6i shows that the recycled material main-

tains an intact crystal structure. This recycling strategy significantly decreases greenhouse gas (GHG) emissions (Figure 6j), offering clear benefits in energy efficiency, environmental impact, and economics.

For elemental extraction from LFP batteries, the main focus is lithium extraction because of its high recycling value^[140,141]. To maximize this extraction efficiency, Fu et al.^[142] developed a novel three-stage leaching system utilizing spent LFP, nitric acid (HNO_3), and oxygen (O_2) with a leaching rate of almost 100%. Furthermore, Wang et al.^[108] demonstrated an electrochemical method (Figure 6k) capable of simultaneously recovering lithium from spent lithium-ion batteries (achieving an extraction efficiency of 93%, Figure 6l) and capturing nitrogen dioxide from exhaust gases, while also generating electrical energy and high-purity lithium nitrate. This process produces CO_2 emissions that are only one-tenth of those from other methods, indicating its potential as a green and sustainable approach (Figure 6m).

Crucially, for LFP cathode materials, the decision between direct recycling and elemental extraction is profoundly influenced by dynamic economic and market factors^[143–145]. Given the relatively low intrinsic value of iron and phosphorus in LFP, the overall recycling economics are predominantly dictated by lithium prices and processing costs^[146,147]. When lithium prices are high, selective lithium extraction can generate robust profit margins, making it the preferred industrial choice. However, under moderate to low lithium price environments, these profit margins shrink or even disappear. In such scenarios, although direct recycling or upcycling offers superior material circularity and a lower carbon footprint, they suffer from initial cost disadvantages. Therefore, the optimal recycling route for LFP is not static; it must be continuously re-evaluated against real-time material degradation states, market prices, policy incentives, and technological maturity^[148].

5.3. Recycling of lithium manganese oxide cathode materials

The degradation of lithium manganese oxide (LMO) cathode material is mainly governed by irreversible structural failure driven by Jahn-Teller distortion and Mn^{3+} disproportionation^[34,57,149,150]. Strategies such as direct recycling and upcycling are commonly used to restore the lattice structure of degraded materials through methods such as solid-state sintering or Joule heating (Figure 7a). For example, Ji et al.^[150] provided a detailed protocol for the direct recycling and upcycling of spent LiMn_2O_4 into cathode materials with future market potential, such as high-voltage $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ or lithium-rich $\text{Li}_{1.2}\text{Ni}_{0.2}\text{Mn}_{0.6}\text{O}_2$. At the microstructural level, this approach effectively repairs lattice collapse in spent materials, with the degraded phases being completely removed (Figure 7b). For materials with more severe damage, various leaching-based recovery methods are applied^[151]. For example, He et al.^[152] reported a low-temperature $(\text{NH}_4)_2\text{SO}_4$ calcination mechanism followed by water leaching to convert LiMnO_4 into soluble sulfates (Figure 7c). Alternatively, spent materials can be directly recycled as catalysts^[153] or repurposed for other battery applications. Zhang et al.^[154] demonstrated the integrated recovery of both the cathode active material and the aluminum foil current collector from spent lithium-manganese batteries to prepare aluminum-doped manganese dioxide for use

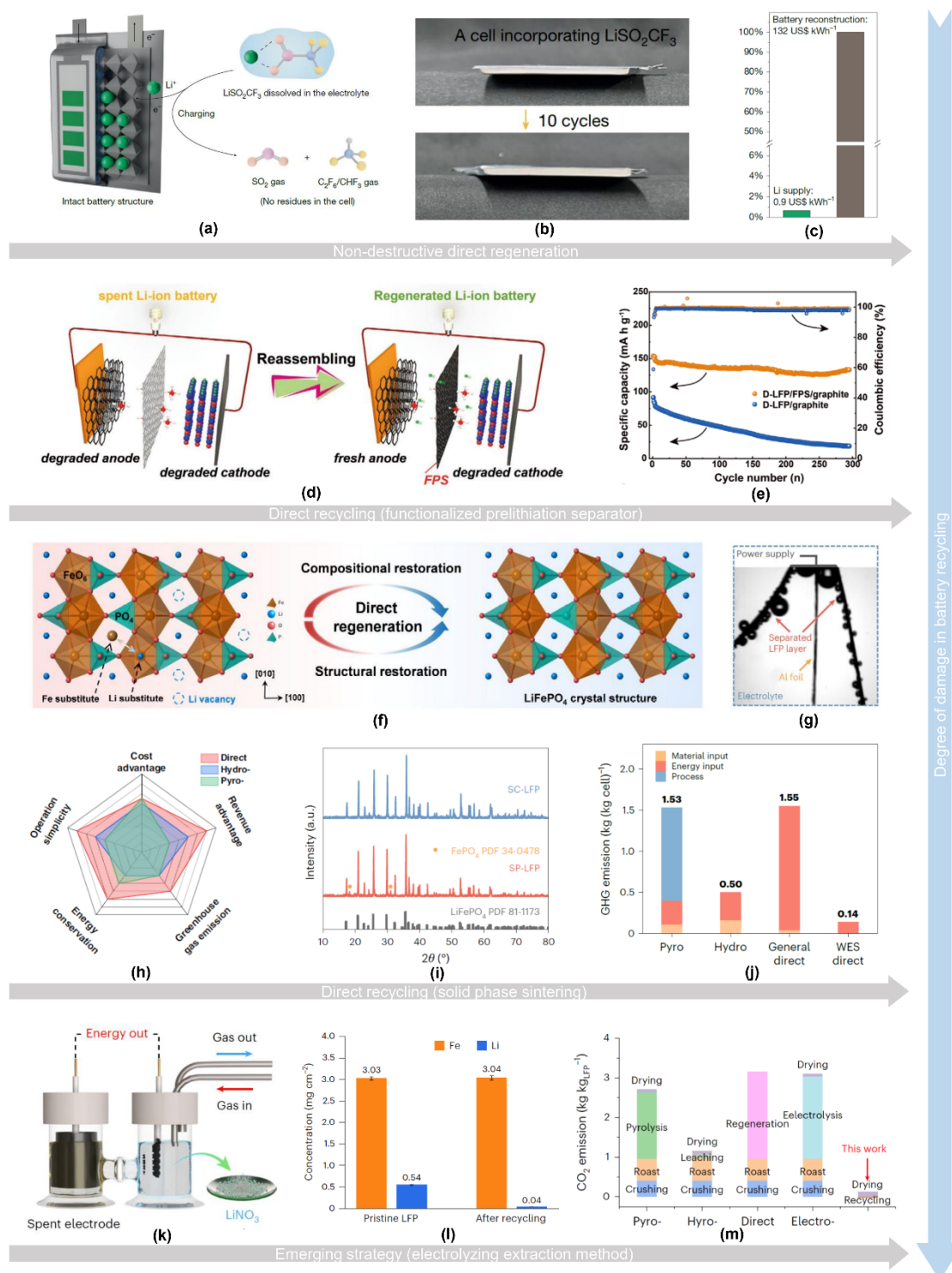


Fig. 6. Recycling of LFP cathode. (a) Schematic of an external Li supply strategy by incorporating an organic LiSO_2CF_3 salt into assembled cells. This replenishes lost lithium inventory without dismantling the cell; (b) Photographs of cells after 10 cycles using the LiSO_2CF_3 additive, showing no swelling or gas residue, confirming the safety and non-destructive nature of the process; (c) Comparison of energy storage costs in power grids; adapted from Ref. [104]. (d) Schematic illustration of the direct recycling strategy based on a functionalized prelithiation separator. This strategy bypasses complex material regeneration by directly compensating for Li loss during subsequent cycling; (e) Cycling stability plots demonstrating that the FPS enables degraded LFP to maintain excellent capacity retention compared to rapid failure with a normal separator; adapted from Ref. [138]. (f-j) Material-level direct recycling: (f) Schematic of the degraded and restored LFP crystal structures; (g) Separation of LFP cathode using the water electrolysis-induced separation method; (h) Radar chart highlighting the advantages of direct recycling over pyro/hydrometallurgy in cost, revenue, and operational simplicity; (i) XRD patterns confirming that the SC-LFP maintains a relatively intact crystal structure; (j) EverBatt analysis quantitatively proving that the WES direct route significantly minimizes GHG emissions; adapted from Ref. [106,107]. (k) Schematic diagram of a scaled-up H-type battery for electrochemical recycling; (l) The drastic drop in lithium content (from 0.54 to 0.04 mg cm^{-2}) while retaining Fe demonstrates highly efficient and selective elemental Li extraction; (m) CO_2 emissions evaluation showing that this emerging electrochemical method drastically reduces the carbon footprint; adapted from Ref. [108].

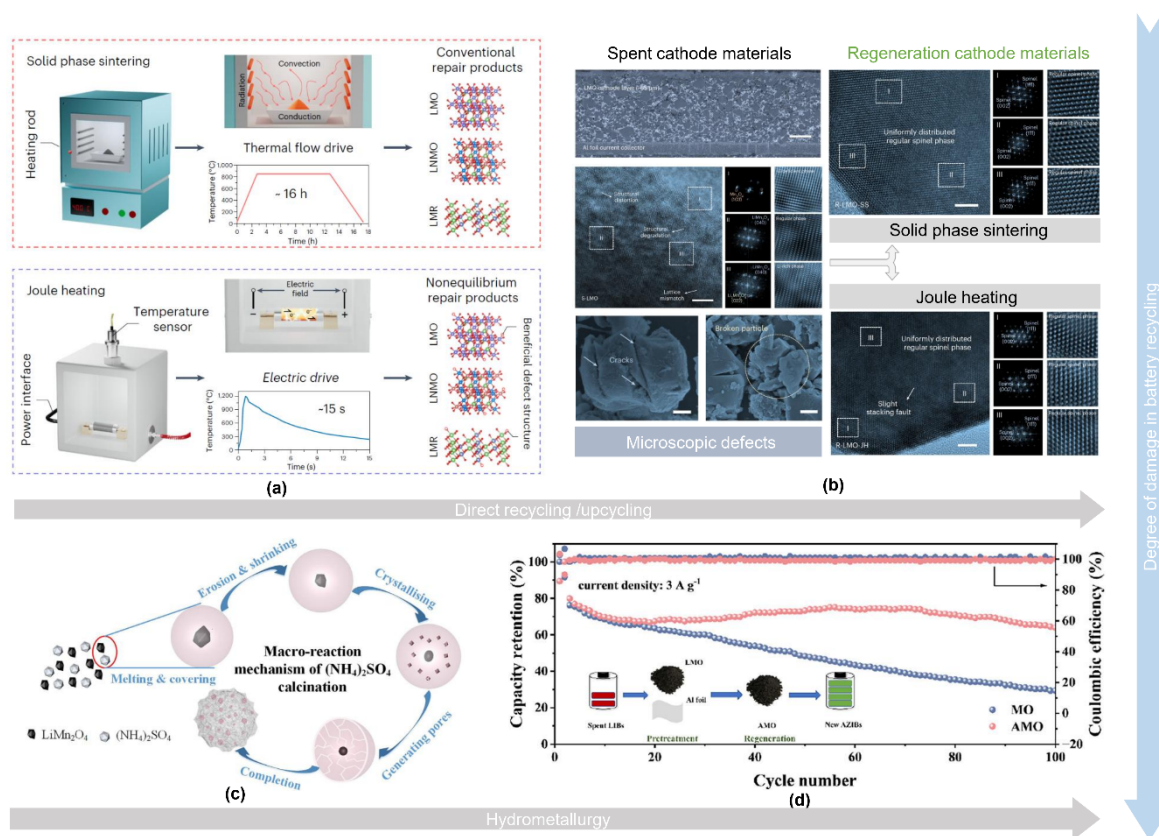


Fig. 7. Recycling of lithium manganese oxide cathode. (a) Mechanistic comparison between conventional solid-phase sintering and joule heating for LMO regeneration; (b) Microscopic image of spent cathode materials and regenerated LiMn_2O_4 cathode materials; adapted from Ref.^[150]. (c) Schematic diagram of low-temperature calcination recovery of waste LiMn_2O_4 using $(\text{NH}_4)_2\text{SO}_4$; adapted from Ref.^[152]. (d) Cycling performance of aqueous zinc batteries prepared from recycled LMO for MO and AMO; adapted from Ref.^[154].

as a cathode material in aqueous zinc-ion batteries (Figure 7d). Furthermore, to address the complexity of failure states in practical recycling scenarios—such as the coexistence of mixed spent LMO and LFP, Chen et al.^[155] developed a novel one-step strategy integrating leaching–sol–gel and subsequent annealing to directly upcycle mixed spent cathodes into high-value lithium manganese iron phosphate (LMFP).

5.4. Industrial-scale implementation challenges

Although laboratory-scale studies have convincingly demonstrated the feasibility of precision recycling, translating these strategies into industrial practice is fundamentally a complex systems-engineering challenge. At the commercial scale, feedstock heterogeneity directly threatens the viability of direct recycling and upcycling, demanding robust automated sorting technologies to manage mixed battery chemistries and physical formats^[59,156]. This is further complicated by stringent impurity control requirements, as direct recycling exhibits exceptionally low tolerance for trace metals (e.g., Cu, Al, Fe) compared to traditional hydrometallurgy^[157,158]. Furthermore, process scalability faces significant engineering hurdles, such as maintaining uniform thermal and thermodynamic conditions in ton-scale reactors to ensure batch-to-batch consistency^[159]. Operationally, safety management must rigorously address thermal runaway risks during dismantling and the handling of toxic gases^[160,161]. Consequently, the technology readiness levels (TRLs) across these pathways vary drastically^[162]; while pyrometallurgy and hydrometallurgy are fully commercial-

ized (TRL 8–9), direct recycling (TRL 4–6), upcycling (TRL 3–5), and non-destructive direct regeneration (TRL 1–3) are still traversing the pilot and proof-of-concept stages. Realizing a truly circular battery economy will therefore require concerted efforts that extend beyond materials science to encompass holistic process engineering, supply-chain logistics, and standard-setting^[163,164].

6. Conclusion and outlook

This review systematically examines the failure mechanisms of cathode materials (layered oxides, lithium iron phosphate, and spinel lithium manganese oxide,) and comprehensively summarizes the mainstream recycling strategies (pyrometallurgy, hydrometallurgy, direct recycling, upcycling and non-destructive direct regeneration) that correspond to these mechanisms. A central thesis is established and supported: the selection of a recycling strategy is a precise matching process determined by the specific failure state of the material. To achieve an efficient and green future for battery recycling, the field must shift from the current rough “processing” paradigm toward a science-informed “precision recycling” paradigm. The core of this paradigm is a closed-loop system built on a deep understanding of failure mechanisms, guided by accurate diagnosis of material states, and supported by the intelligent matching of multiple technological pathways.

Looking toward the technological frontier of “AI+”^[165–168], paradigm shifts in battery cathode material recycling will

strongly depend on the integration of digital intelligence technologies, represented by multimodal AI and digital twins, with physical recycling processes. This integration will promote the establishment of a closed-loop system involving “non-destructive/minimally invasive failure analysis—intelligent decision-making—precision recycling,” enabling a fundamental shift from an “experience-driven” approach to a “data- and model-driven” approach (Figure 8). However, the efficacy of AI-driven paradigms inherently hinges on robust, large-scale datasets. Herein, we issue a call to action for future researchers to systematically document detailed, quantitative cathode degradation metrics (e.g., precise lithium inventory loss ratios, structural phase transition percentages, and transition-metal

dissolution concentrations) in forthcoming studies. Such standardized data repositories will serve as the indispensable bedrock for realizing AI-enabled, closed-loop precision recycling designs. Meanwhile, the application of artificial intelligence can enable adaptive, data-informed decision-making throughout the battery life cycle, effectively overcoming the critical barrier caused by the lack of reliable data regarding the state of retired batteries. AI technologies are capable of undertaking diverse tasks tailored to different stages of the battery life cycle. By covering key evaluation indicators—including the state of charge (SOC), state of health (SOH), and remaining useful life (RUL)—these algorithms realize the precise deduction of unobservable internal states within the cells^[169].

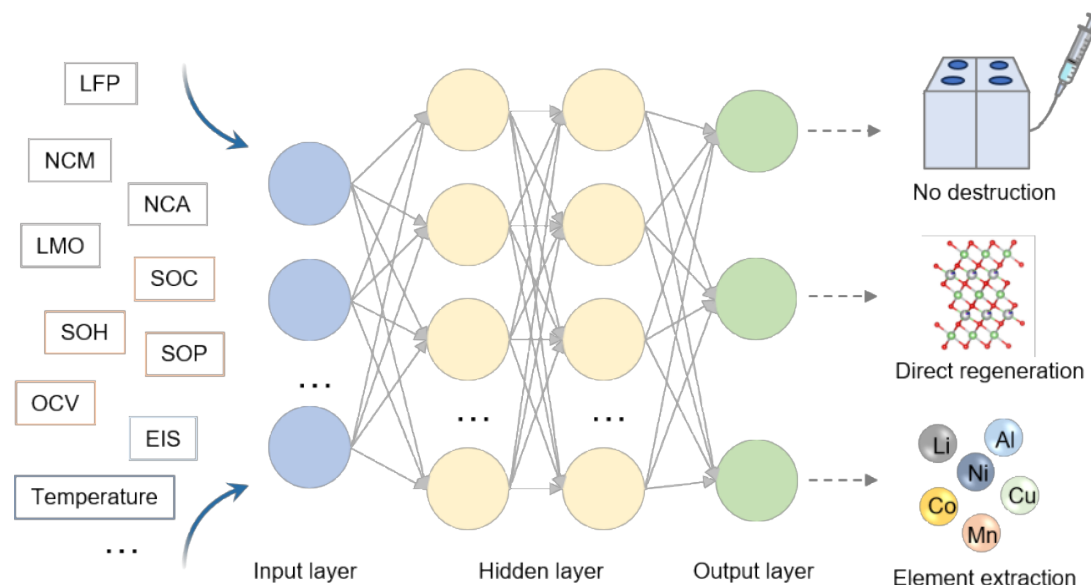


Fig. 8. Schematic illustration of data-driven precision recycling. Abbreviations used in the figure: SOC (State of Charge), SOH (State of Health), SOP (State of Power), OCV (Open Circuit Voltage), EIS (Electrochemical Impedance Spectroscopy).

(1) Multimodal AI-enabled intelligent failure diagnostics and prognostics. By integrating data from various non-destructive or minimally invasive sensing techniques (e.g., electron microscopy, electrochemical impedance spectroscopy, charge-discharge profiles), multimodal AI models can perform rapid online assessment of battery packs, modules, or individual cells. Through deep learning of parameters such as remaining capacity and internal resistance, and by extracting correlative features from large historical failure datasets, AI can inversely infer the type and severity of internal cathode material degradation, thereby achieving intelligent diagnostics that connect macroscopic performance with microscopic mechanisms.

(2) Digital twin-driven precise process design and dynamic optimization. For a defined recycling strategy, a digital twin model can be further constructed. This model acts as a virtual mirror of the physical plant, integrating thermodynamics, kinetics, fluid dynamics, and chemical reaction models, and can be continuously updated with real-time sensor data^[170–172]. It enables the evaluation of economic and environmental feasibility for commercial-scale recycling processes before physical implementation.

To fully realize precision recycling, several key scientific challenges must be systematically addressed. These include

the standardization of battery-health diagnostics, a deeper mechanistic understanding of regeneration limits, the continuous development of low-carbon regeneration processes, and the ultimate establishment of closed-loop industrial ecosystems. In particular, the deep integration of artificial intelligence with physical recycling facilities for the rapid and accurate diagnosis of spent batteries, together with the automated matching and optimization of recycling pathways, will be crucial for achieving efficient and sustainable battery recycling in the future.

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Conflict of interest

The authors declare no competing financial interest.

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