

From Battery Ageing to Water Treatment: Catalytic Upcycling of Spent Lithium-ion Battery Electrodes

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Abstract: The growing stock of spent lithium-ion batteries and the increasing demand for advanced water treatment represent two closely connected sustainability challenges. Conventional battery recycling primarily targets the recovery of lithium and transition metals as purified salts or regenerated electrode materials. These routes often dismantle or overlook the chemically complex structures retained within aged electrodes. Catalytic upcycling offers an alternative pathway by redirecting spent cathodes and anodes towards contaminant degradation, in situ oxidant production and electrochemical water treatment. Spent cathodes contain redox-active Co, Ni, Mn and Fe species, whereas spent anodes provide conductive graphite frameworks with tunable defects and surface functionalities. Electrochemical ageing may further generate lithium-deficient domains, oxygen vacancies, reconstructed interfaces and microcracks, some of which can act as a form of service-induced pre-activation. In this Mini Review, we connect these ageing-derived characteristics with the conversion and catalytic functions of representative spent electrode materials. We compare direct reuse, mild activation and phase-reconstruction strategies, and examine their applications in Fenton-like reactions, persulfate activation and electrocatalytic water treatment. We argue that catalytic activity alone is insufficient to establish sustainable upcycling. Conversion intensity, catalyst lifetime, metal release, toxicity of transformation products, and performance in realistic water matrices must also be considered. Finally, we outline opportunities for low-energy conversion, safer catalyst design, continuous-flow treatment, and data-driven optimization. This framework links battery ageing with catalytic function while defining the conditions required for environmentally credible water treatment applications.

Keywords: spent lithium-ion battery electrodes; catalytic upcycling; peroxymonosulfate activation; H₂O₂ electrosynthesis; electro-Fenton treatment



1. Introduction

The expanding deployment of lithium-ion batteries in portable electronics, electric vehicles, and stationary energy-storage systems is creating a rapidly growing and chemically diverse stream of spent batteries [1–3]. In parallel, the persistence of organic contaminants in municipal and industrial effluents continues to motivate the development of efficient advanced oxidation and electrochemical treatment processes [4,5]. Conventional recycling routes primarily recover lithium and transition metals as purified salts or regenerate electrode materials for renewed battery use. Although these strategies are essential for critical-material recovery, they commonly require extensive dismantling, separation, and reprocessing, and may overlook the chemically active features retained in aged electrodes [1–3]. Catalytic upcycling provides a complementary perspective by redirecting spent lithium-ion battery electrodes towards water-treatment functions, including oxidant activation, in situ H_2O_2 production, and electro-Fenton treatment (Figure 1). This opportunity arises from the complementary properties of the two electrode streams: redox-active transition metals in spent cathodes and conductive, defect-rich carbon in spent anodes. Service-related structural and interfacial changes may further modify site accessibility and electron-transfer kinetics, allowing spent electrodes to be regarded as functionally altered catalytic feedstocks rather than only sources of recoverable metals [4].

Growing evidence supports the use of spent cathodes, anodes and black mass in peroxide activation, persulfate-based oxidation and electrochemical water treatment [6–12]. Directly reused or mildly activated LiCoO_2 , Ni-Co-Mn oxides, LiMn_2O_4 and LiFePO_4 can activate peroxymonosulfate, H_2O_2 or peracetic acid through transition-metal redox cycling and defect-mediated electron transfer [6–8,11,12]. In parallel, spent graphite can be tailored for the two-electron oxygen reduction reaction, H_2O_2 electrosynthesis, electro-Fenton treatment, and as a conductive support for metal-based catalysts. More intensive chemical conversion and material-reconstruction routes further enable the preparation of oxides, hydroxides, ferrites, and metal-carbon composites with tunable composition and morphology [3,8]. However, the reported literature remains fragmented across battery chemistries, conversion methods and reaction systems. Many studies emphasize pollutant removal in single-compound batch tests, whereas the effects of feedstock variability, residual binders and electrolyte species, metal leaching, catalyst ageing, transformation-product toxicity and realistic water matrices are less consistently addressed. High catalytic activity alone also does not establish that an upcycling route is environmentally preferable to conventional metal recovery, electrode regeneration or catalyst synthesis from virgin precursors. A meaningful assessment must therefore integrate conversion intensity, reagent and energy inputs, catalyst lifetime, metal mass balance, treatment performance in complex water matrices, and life-cycle environmental and economic implications [4,12].

This review develops an ageing-to-function framework for understanding the catalytic upcycling of spent lithium-ion battery electrodes for water treatment. It first relates the service-induced structural and interfacial characteristics of representative cathode and anode materials to their catalytic potential. It then compares direct reuse and mild activation with chemical conversion and material reconstruction, highlighting the trade-offs between preserving ageing-derived active structures and achieving compositional control, reproducibility and stability. The subsequent sections synthesize water-treatment applications in peroxymonosulfate activation, H_2O_2 electrosynthesis, electro-Fenton treatment and other emerging catalytic processes. Particular attention is given to the interconnected challenges of feedstock heterogeneity, metal release, catalyst durability, real-wastewater performance, and environmental and economic feasibility. Finally, future directions are proposed for low-energy conversion, safer catalyst design, continuous-flow operation, and data-driven optimization. Rather than presenting catalytic upcycling as a universal substitute for established battery-recycling routes, this review identifies the conditions under which it can provide a credible and environmentally defensible pathway for linking battery ageing with water-pollution treatment.

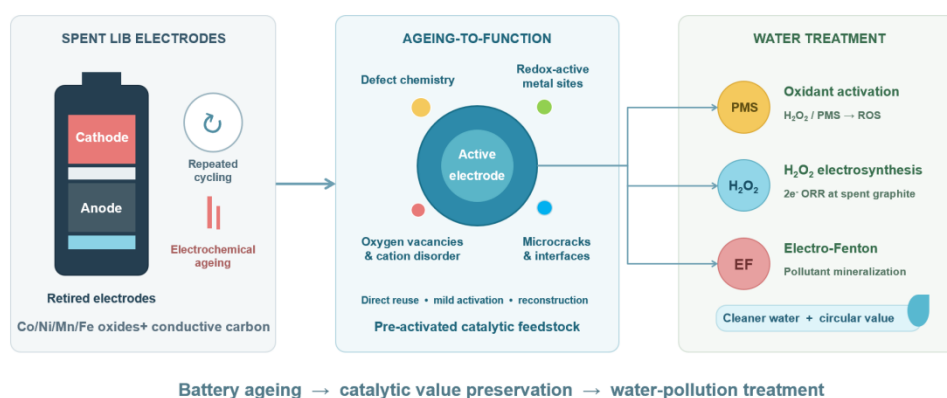


Figure 1. Ageing-to-function framework for LIB electrode upcycling in water treatment.

2. Ageing-Derived Catalytic Attributes of Spent Lithium-ion Battery Electrodes

The catalytic potential of spent lithium-ion battery electrodes arises from non-equilibrium structures and interfacial features generated during extended service. This section focuses on the resulting structure-property relationships, whereas application-specific reaction conditions and performance metrics are compared in Section 4. Spent cathodes provide redox-active transition-metal sites, while spent graphite retains a conductive carbon framework with tunable defects and surface functionalities. Battery ageing can further generate lithium-deficient domains, cation disorder, oxygen vacancies, reconstructed interfaces and microcracks that modify site accessibility, adsorption and electron-transfer kinetics [4].

2.1. Cathode-Derived Active Motifs Across Chemistries

Across lithium cobalt oxide (LCO), lithium nickel-cobalt-manganese oxide (NCM), lithium manganese oxide (LMO) and lithium iron phosphate (LFP) cathodes, catalytic behavior reflects the combined effects of transition-metal composition and ageing-derived structural changes. Lithium loss, cation disorder, oxygen vacancies, surface reconstruction and micro cracking can expose coordinatively unsaturated sites and shorten diffusion pathways. However, the consequences of these features are chemistry dependent because each cathode family provides a different redox environment [4–7,13–15].

LCO primarily provides $\text{Co}^{2+}/\text{Co}^{3+}$ redox centers for peroxide activation, whereas NCM combines Ni, Co and Mn sites whose coupled valence changes can broaden redox pathways but also create composition-dependent activity-stability trade-offs. In LMO, the $\text{Mn}^{3+}/\text{Mn}^{4+}$ couple supports oxidant activation, although Jahn-Teller distortion and Mn dissolution constrain long-term use. LFP instead supplies Fe-centered Fenton-like activity and offers a comparatively low-toxicity, earth-abundant metal basis. These distinctions define the expected catalytic roles of the four cathode families; their reported treatment performance is evaluated in Section 4 [5–7,14,15].

Ageing-derived defects are beneficial only when they remain accessible without causing excessive structural collapse, loss of conductivity or accelerated metal release. The relevant design question is therefore not whether a spent cathode contains more defects, but which defects are accessible, stable and compatible with the intended catalytic pathway. This conditional view also helps distinguish transferable structure-activity relationships from effects that are specific to a particular battery chemistry or waste batch.

2.2. Spent Graphite Anodes as Defect-Rich Carbon Platforms

Spent graphite differs from cathode materials because its principal catalytic functions arise from carbon defects, surface oxygen groups and retained electrical conductivity. Repeated Li^+ intercalation and deintercalation, solid-electrolyte interphase evolution and particle fracture can generate edge sites, vacancies, turbostratic domains and micro cracks. These features alter O_2 adsorption and electron transfer, providing a material basis for the two-electron oxygen reduction reaction and related electrochemical water-treatment processes [8,9,16].

Post-use treatment determines how these ageing-derived attributes are expressed. Acidic or oxidative treatment can expose or introduce carboxyl, hydroxyl and ether groups, whereas stronger purification can remove residual metals and substantially change the surface chemistry. Spent graphite may therefore function as an H_2O_2 -generating cathode, an electro-Fenton interface or a conductive support for metal-based catalysts. The corresponding application metrics and case studies are retained in Section 4 to avoid repeating them here [8,9,17,18].

2.3. Cross-Electrode Design Principles from Ageing to Catalytic Function

Across cathodes and anodes, the ageing-to-function relationship is primarily kinetic and interfacial: microcracks can expose previously inaccessible sites, vacancies can alter adsorption configurations, multivalent metal centers can mediate redox cycling, and defective carbon can facilitate interfacial charge transfer. Catalysts do not alter the Gibbs free-energy change or equilibrium position of the overall reaction; instead, ageing-derived features may lower activation barriers and alter reaction pathways or selectivity. These effects are chemistry- and condition-dependent, and their practical value remains contingent on feedstock consistency, structural integrity and control of metal or particulate release. This framework separates the material basis of catalytic activity discussed in Section 2 from the conversion routes in Section 3 and the application-specific evidence in Section 4.

3. Catalytic Upcycling Strategies: Direct Preservation Versus Material Reconstruction

Processes for preparing water-treatment catalysts from spent electrodes can be grouped into two routes: direct reuse or mild activation, which preserves the native electrode architecture through low-damage pretreatment; and

chemical conversion or material reconstruction, which reorganizes the material through leaching, mechanochemistry, microwave treatment, doping or related methods to control composition and morphology.

3.1. Direct Reuse and Mild Electrode Activation

Direct reuse and mild activation employ disassembly, cleaning, acid/alkali etching, low-temperature heat treatment or particle-size screening without fully dismantling the electrode architecture. These steps remove impurities or tune accessible sites while limiting compositional reconstruction.

3.1.1. Separation, Cleaning and Interfacial Defect Regulation

After thorough short-circuit discharge of spent lithium-ion batteries, mechanical crushing combined with manual sorting is employed to separate the cathode and anode sheets, followed by rinsing with N-methyl-2-pyrrolidone organic solvent to remove residual electrolyte and polyvinylidene fluoride (PVDF) binder from the electrode surface. The electrolyte and organic binder coat the metal active sites, directly blocking the electron transfer pathways for catalytic oxidation; the cleaning process eliminates the shielding effect of impurities and prevents the decomposition of organic contaminants into toxic byproducts during water treatment. Wang et al. [6] demonstrated that LiMn_2O_4 electrode sheets after simple cleaning and impurity removal, without the need for deep purification, can be directly utilized as recyclable heterogeneous peroxymonosulfate (PMS) catalysts.

Controlled acid/alkali etching is the core modification method for the simple activation of spent electrodes, enabling precise regulation of interfacial structure and functional groups. Through the differentiated effects of acid washing and alkaline washing, it can simultaneously achieve defect construction in the cathode, functional group reconstruction on the anode, and impurity removal, serving as a key process for cost-effectively enhancing the water-treatment catalytic performance of spent electrodes. For graphite anodes from retired batteries, acid washing can selectively etch inert sites at carbon edges, directionally enriching oxygen-containing active functional groups such as carboxyl, hydroxyl, and C-O-C, effectively reducing the energy barrier for the two-electron oxygen reduction reaction and promoting the efficient directional conversion of O_2 to H_2O_2 making it suitable for electro-Fenton water treatment systems. Cao et al. [8] investigated the electro-Fenton performance of three types of spent graphite anodes: raw powder (RP), acid-leached (AL), and acid-alkali leached (AAL). Acid leaching significantly increased the carboxyl group content (35.83% for AL vs. only 7% for AAL), which enhanced the interfacial adsorption of Fe ions on the electrode surface, established a heterogeneous iron redox cycle, and accelerated the Fenton reaction. The AL electrode achieved complete BPA degradation within 70 min and 87.4% Chemical Oxygen Demand (COD) removal within 240 min, with no performance decay after 10 reuse cycles, outperforming conventional carbon felt. Although alkali leaching effectively removed metal impurities, excessive reduction of carboxyl groups compromised the degradation efficiency.

Low-temperature heat treatment ranging from 300 to 500 °C is recognized as an eco-friendly mild activation strategy for spent electrode catalysts, which has been systematically validated by Lin et al. [19] via nitrogen calcination of waste LiCoO_2 cathodes at 500 °C. Such mild thermal treatment fully retains the inherent metal-carbon composite framework and layered crystal structure formed during battery cycling; moderate thermal stimulation expands lattice microcracks and enriches surface oxygen/metal vacancies simultaneously, which optimizes interfacial charge transfer kinetics, lowers electron transfer resistance in catalytic cycles, and greatly elevates the mineralization efficiency of organic contaminants. Unlike high-temperature calcination (>800 °C) that causes severe metal agglomeration, erases service-generated intrinsic defects and destroys conductive carbon networks, low-temperature thermal pretreatment achieves a favorable trade-off between catalytic activity, processing cost and operational controllability.

In summary, a series of mild pretreatment methods including disassembly and cleaning, controlled acid/alkali etching, low-temperature thermal activation, and particle size classification can, without destroying the native metal-carbon composite structure of spent electrodes and while preserving the natural defects derived from battery cycling, precisely optimize surface functional group distribution, amplify active sites, and reduce interfacial mass transfer impedance, thereby achieving efficient enhancement of the intrinsic catalytic activity of spent electrodes. After this short-process mild modification, the spent electrodes and black powder materials, without the need for complex steps such as deep metal leaching, recrystallization, or material reconstruction, can be directly applied as heterogeneous PMS activators and electro-Fenton cathodes in various organic wastewater treatment systems. The entire process features low chemical and energy consumption, simple unit operations, strong engineering feasibility, and a small carbon footprint, representing the most cost-effective, highly adaptable, and atom-economical shallow-level catalytic resourceization approach for retired lithium-ion battery electrodes at the current stage.

3.1.2. Advantages: Short Process Chains and Defect Preservation

The principal engineering advantage of direct reuse and mild activation is the preservation of the original electrode architecture with fewer conversion steps. This can retain delithiation vacancies, interlayer microcracks, metal-carbon interfaces and the conductive graphite framework without separately synthesizing a support. Nevertheless, solvent washing, acid or alkali treatment, repeated rinsing, drying and thermal activation up to approximately 500 °C may still dominate the environmental burden. Claims of lower cost or emissions therefore require measured electricity, heat, water and reagent inventories, conversion yield, catalyst lifetime and end-of-life recovery, rather than qualitative labels such as green or low-energy.

3.1.3. Limitations: Feedstock Heterogeneity and Constrained Compositional Control

Although the simple activation process offers numerous advantages including low cost, low energy consumption, and easy implementation, this technical system still suffers from unavoidable inherent limitations that greatly constrain its large-scale engineering applications [20]. Retired lithium-ion batteries encompass multiple cathode systems including LCO, NCM, LMO, and LFP, with significant differences in metal element molar ratios across different systems. In addition, variations in battery cycle life, state of charge, and thermal failure modes further alter the metal valence distribution and defect density of the electrodes, resulting in strong intrinsic heterogeneity in industrial mixed black powder feedstocks and prominent batch-to-batch fluctuations in catalytic performance. Ji et al. [21] highlighted this industrial recycling bottleneck and compared conventional recovery routes with their deep eutectic solvent (DES) upgrading strategy. Pyrometallurgical and hydrometallurgical recycling can generate secondary pollution and low-value Fe/Mn products. Direct regeneration may not sufficiently rearrange transition metals because LFP and LMO have mismatched crystal structures. The CHCl_3 -oxalate DES process requires prolonged leaching at 110 °C and additional water-dilution steps. Its reported raw-material cost was US\$2.01 kg⁻¹ of feedstock, the highest among the mainstream recycling technologies considered. Moreover, the process has been demonstrated only at gram scale and has limited adaptability to mixed cathode waste containing NCM and other impurities. It also does not establish a complete closed-loop pathway for the secondary recycling of regenerated lithium-iron-manganese phosphate (LFMP). These limitations leave feedstock heterogeneity, reagent cost and industrial scale-up as unresolved barriers to practical deployment.

3.2. Chemical Conversion and Catalyst Reconstruction

Chemical conversion and material reconstruction comprise deep-deconstruction routes that transform spent electrodes through component recovery, controlled precursor formation and reconstruction into functional water-treatment catalysts. Hydrometallurgical leaching, mechanochemical processing, sol-gel synthesis and hydrothermal treatment can separate or redistribute metal and graphite components while controlling composition, morphology and interfacial defects. Compared with direct reuse, these routes offer greater compositional control but require more processing steps and chemical or energy inputs.

3.2.1. Reconstruction Routes and Physicochemical Regulation

Hydrometallurgical leaching coupled with material reconstruction represents a mainstream upcycling strategy to transform waste lithium-ion cathode materials into heterogeneous PMS activators. In this integrated technical route, mineral acids or green deep eutectic solvents (DES) are adopted as leaching media to extract target metals from spent cathodes; subsequent solid-liquid separation removes Al foil fragments, PVDF binder and graphite impurities to obtain purified metal-bearing solution. By adjusting solution pH or regulating chelation reactions, material reconstruction can be precisely controlled to fabricate precursors with abundant redox-active sites. Xu et al. [22] adopted citric acid-assisted sol-gel reconstruction based on acid leachate of waste NCM cathodes, and the derived ternary SG-NCM catalysts exhibited a 4-fold higher PMS activation kinetic constant than unprocessed spent electrode scraps. The strong orbital coupling between multivalent Co and Mn generates synergistic catalytic centers, facilitating the production of singlet oxygen and sulfate radicals to achieve efficient mineralization of organic pollutants.

Solvent-free mechanochemical solid-phase reconstruction relies on the mechanical force of high-energy ball milling to break metal-oxygen bonds, induce lattice disordering, and simultaneously generate abundant oxygen vacancies and valence distortions, without the need for large quantities of acid/alkali solvents throughout the process, representing a solid-phase green reconstruction route. This is well exemplified by the work of Chen et al. [17], who employed mechanochemical ball milling pretreatment to process spent lithium-ion battery anode scraps for fabricating graphite-supported zero-valent iron-copper bimetallic catalysts (ZVI-Cu/C) coupled with mill scale

steel waste. During this ball milling mechanochemical activation process, intense mechanical collision, friction and extrusion completely stripped the graphite coating from copper foil and shattered the layered anode composite structure, achieving uniform mixing of graphite and copper microparticles while introducing structural defects and lattice disorder on carbon matrix. After subsequent carbothermal reduction at 1000 °C under nitrogen atmosphere, the iron oxides in mill scale were reduced to zero-valent iron, and the mechanically induced defects and vacancies on graphite support further promoted interfacial electron transfer between ZVI and Cu bimetallic sites. This mechanochemical pretreatment strategy realized valence transformation of iron species ($\text{Fe}^{3+}/\text{Fe}^{2+}/\text{Fe}^0$ cycling) and produced abundant surface oxygen vacancies on the bimetallic composite, which significantly boosted both reductive dechlorination and heterogeneous Fenton oxidation catalytic performance for refractory 4-chlorophenol water pollutant degradation.

Microwave-assisted field-enhanced leaching was adopted to strengthen hydrometallurgical metal recovery from retired lithium-ion battery cathodes. Kotkar et al. [23] introduced microwave-enhanced sulfuric acid leaching with H_2O_2 as a reductant. Under optimized conditions (60 °C, 30 min, 0.5 M H_2SO_4 , 50 W microwave power, and 3 vol% H_2O_2), nearly complete leaching of Li, Co, Ni and Mn was achieved, with leaching efficiencies of Li: 89.07%, Co: 94.57%, Ni: 94.25%, Mn: 100%. After leaching, NaOH was added at ambient temperature for coprecipitation to harvest $\text{Co}(\text{OH})_2$ doped with trace Ni/Mn, which acts as high-performance electrocatalyst for oxygen evolution reaction (OER). Though microwave treatment only worked on leaching unit, the recycled hydroxide catalyst delivered far lower charge transfer resistance (5.5 Ω) than commercial $\text{Co}(\text{OH})_2$ (14.6 Ω) and benchmark IrO_2 electrocatalysts (20.8 Ω), demonstrating accelerated interfacial electron transfer and superior OER reaction kinetics.

Phosphidation treatment of metal hydroxide or oxide precursors serves as an effective anion doping strategy, introducing P^{3-} species to modulate the electronic structure and d-band center position of transition metals. This modification facilitates the construction of metal phosphide or sulfide active sites with enhanced intrinsic activity, while also promoting charge transfer kinetics and improving the activation capability toward oxidants. In a representative study, Wang et al. [24] successfully converted leachate from spent NCM cathodes into multimetallic phosphide catalysts. Through P doping, the electron cloud distribution around Co, Ni, and Mn was effectively regulated, leading to optimized intermetallic synergistic effects. As a result, the obtained phosphide materials demonstrated significantly superior OER performance compared to their hydroxide counterparts. Notably, the phosphidation treatment also played a crucial role in stabilizing the metal species, substantially mitigating the risk of metal ion dissolution during electrochemical operation.

3.2.2. Advantages: Controllable Composition and Tailored Catalytic Architectures

The core advantages of the chemical reconstruction route are manifested in four aspects. Metal composition is highly controllable, as leaching and purification achieve complete removal of impurities, allowing on-demand regulation of Ni/Co/Mn/Fe molar ratios, eliminating batch-to-batch performance variations in industrial black powder from the source, and resulting in catalytic materials with excellent reproducibility. Microstructures can be precisely engineered artificially, enabling the directed construction of special morphologies such as ultrathin nanosheets, porous hollow structures, heterojunctions, phosphides, or sulfides, generating high-density unsaturated active sites, with the upper limit of catalytic performance far exceeding that of native simply activated materials. Impurities can be thoroughly separated, as leaching and impurity removal processes completely eliminate PVDF, Cu/Al debris, and electrolyte residues, significantly suppressing metal leaching during catalytic operation, facilitating the distinction of pure solid-phase heterogeneous catalytic contributions, and avoiding homogeneous leaching interference. The application scope is broader, as the route enables the directed preparation of multiple types of functional catalytic materials for electro-Fenton, persulfate activation, photocatalysis, electroreductive denitrification, and heavy metal adsorption, making it suitable for challenging and complex industrial wastewater treatment systems.

3.2.3. Limitations: Multistep Processing and Environmental Burdens

Chemical reconstruction routes also suffer from inherent limitations. The process entails a lengthy workflow consisting of multiple unit operations, such as leaching, impurity removal, precipitation, thermal treatment and doping modification. A full set of hydrometallurgical equipment requires considerable capital investment, hindering the practical deployment of small- and medium-scale resource recovery projects. Large quantities of acids, alkalis, reducing agents, as well as phosphating and sulfiding reagents are consumed throughout the process, concurrently producing massive amounts of high-salinity acidic wastewater. Subsequent wastewater treatment via neutralization and membrane separation greatly diminishes the environmental merits of solid waste recycling. In

addition, substantial separation loss occurs for the graphitic carbon substrate. Graphite dissolves together with target metals during hydrometallurgical leaching, resulting in the inefficient utilization of carbon resources.

4. Water-Treatment Applications of Spent-Electrode-Derived Catalysts

Building on the structure-property framework established in Section 2, this section focuses on application-specific evidence rather than repeating material descriptions. The literature is organized by treatment platform: cathode-derived oxidant activation, graphite-enabled H_2O_2 electrosynthesis and electro-Fenton treatment, and emerging pathways such as photocatalysis and reductive dechlorination. Within each platform, studies are compared using reaction conditions, treatment performance, mechanistic assignments, stability and reported metal release where available.

Because Co, Ni, Mn and Fe released from spent electrodes can also activate oxidants in solution, the mechanistic labels used below require caution. A heterogeneous surface pathway is considered well supported only when catalytic activity remains associated with the retained solid and cannot be reproduced by dissolved-metal controls at the measured leachate concentrations. Where time-resolved leaching, filtrate activity and colloid separation were not reported, the surface assignment is treated as provisional rather than definitive.

4.1. Oxidant Activation by Cathode-Derived Catalysts: PMS as a Model Platform

Among these platforms, peroxymonosulfate activation has the largest comparative evidence base. The subsections below therefore compare LCO, NCM, LMO, LFP and mixed black mass using their reported treatment performance and operational constraints, while their underlying ageing-derived attributes are discussed in Section 2.

4.1.1. LiCoO₂-Derived Catalysts: Cobalt-Mediated Radical and Non-Radical Pathways

LCO is among the most frequently studied spent cathodes for PMS activation. Its reported activity is commonly attributed to interfacial electron transfer involving Co^{2+} and Co^{3+} . Zhao et al. [5] first used recovered LCO cathode material (containing 94% LiCoO_2 and 6% Co_3O_4) directly as a heterogeneous catalyst for PMS activation to remove antibiotics from wastewater. Their mechanistic analysis indicated that $\text{Co}^{2+}/\text{Co}^{3+}$ redox cycling facilitates PMS activation, while sulfate radicals ($\text{SO}_4^{\cdot-}$) and non-radical singlet oxygen ($^1\text{O}_2$) were identified as the principal reactive species contributing to levofloxacin degradation under the tested conditions. Under optimized conditions (catalyst 0.4 g L^{-1} , PMS 0.5 mM), the levofloxacin removal reached 94% within 60 min, with a pseudo-first-order rate constant of 0.07597 min^{-1} . The catalyst exhibited excellent activity over a wide pH range of 3–9 and still maintained 80% degradation efficiency after four cycles. Moving beyond the direct reuse of recovered LCO, Dang et al. [7] introduced oxygen and metal vacancies into spent LCO through mechanochemical activation, thereby enhancing electron-transfer kinetics and PMS affinity. They found that superoxide radicals ($\text{O}_2^{\cdot-}$) generated during PMS oxidation can break Li-O bonds to achieve 96.6% lithium leaching, while simultaneously inducing partial reduction of Co^{3+} to Co^{2+} accompanied by the formation of oxygen vacancies. Density functional theory (DFT) calculations indicated shifts in the surface charge density and d-band center of the obtained product (Figure 2a), together with a lower calculated PMS adsorption energy than that of pristine LCO. Guo et al. [10] proposed a cascade utilization strategy for spent LCO electrode sheets, first using them as PMS catalysts to degrade organic pollutants, then recovering valuable metals from the spent catalyst with deep eutectic solvents. Compared with pristine LiCoO_2 , the spent cathode sheets exhibited higher catalytic performance. The authors associated this improvement with vacancies and electronic-structure changes generated during repeated de-lithiation; the material achieved over 98% removal of o-phenylphenol within 60 min and maintained >95% removal after 10 cycles.

LiCoO_2 extracted from spent mobile-phone batteries can also effectively activate peracetic acid (PAA): Lin et al. [19] directly used uncrushed spent LCO electrode sheets as PMS catalysts; under conditions of 0.2 g L^{-1} catalyst and 2 mM PMS, the degradation of benzalkonium chloride exceeded 99% within 40 min, with a rate constant of 0.149 min^{-1} , 1.9 times higher than that of fresh Co_3O_4 .

4.1.2. NCM-Derived Catalysts: Multimetal Redox Synergy and Immobilized Catalysis

The multi-metal synergy of Ni, Co and Mn in ternary cathode materials is also fully reflected in PMS activation. Zhao et al. [25] used spent NCM ($\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$) cathode material to prepare NCM-650 catalyst via simple calcination, achieving 100% removal of Rhodamine B (RhB) within 25 min in PMS activation, with a rate constant of 0.179 min^{-1} . Mechanistic studies revealed that the redox cycles of $\text{Co}^{2+}/\text{Co}^{3+}$ and $\text{Mn}^{2+}/\text{Mn}^{3+}/\text{Mn}^{4+}$ on the catalyst surface dominate PMS activation, and $\text{SO}_4^{\cdot-}$, $\cdot\text{OH}$, $\text{O}_2^{\cdot-}$ and $^1\text{O}_2$ all participated in RhB degradation,

with $^1\text{O}_2$ playing the dominant role. The catalyst exhibited excellent activity over a wide pH range (4–10) and retained 98.2% RhB removal after four cycles. Dang et al. [26] treated spent NCM material by electrochemical oxidation to simultaneously recover lithium and obtain a delithiated NCM catalyst with abundant oxygen vacancies (Figure 2b). This catalyst showed excellent performance in PMS-activated degradation of sulfamethoxazole, with 100% removal within 30 min and sustained high activity after five cycles. Liang et al. [27] synthesized magnetic CoFe_2O_4 catalyst from spent ternary LIBs; by regulating the precipitation separation process with organic acids, they achieved efficient separation of Li and Co (Li recovery >96%, Co precipitation efficiency >99%). This catalyst achieved 94% removal of bisphenol A within 25 min in PMS activation, with a rate constant of 0.119 min^{-1} , 1.7 times that of pure CoFe_2O_4 , and could be easily recovered by an external magnetic field. It was found that surface-bound $\text{SO}_4^{\cdot-}$ was the main active species; the higher Co(II)/Co(III) and Fe(II)/Fe(III) ratios, larger specific surface area, and more acidic sites together contributed to the excellent catalytic performance. Xu et al. [22] used a citric-acid-assisted sol-gel method to synthesize SG-NCM400 catalyst from spent NCM leachate, with a specific surface area of $85.6\text{ m}^2\text{ g}^{-1}$. The catalyst achieved over 90% removal of methylene blue within 30 min in PMS activation; $^1\text{O}_2$ was the dominant active species, while $\text{SO}_4^{\cdot-}$, $\cdot\text{OH}$ and $\text{O}_2^{\cdot-}$ also participated. Hu et al. [28] synthesized $\text{CoFe}_2\text{O}_4@\text{CNTs}$ composite catalyst from spent ternary LIBs; the introduction of carbon nanotubes effectively improved the dispersion and electron-transfer ability of CoFe_2O_4 . In PMS-activated degradation of chloroquine phosphate, $\text{CoFe}_2\text{O}_4@4.4\%\text{CNTs}$ achieved 98.7% removal within 25 min, with a rate constant of 0.215 min^{-1} , far exceeding that of pure CoFe_2O_4 . Radical quenching experiments and EPR analysis indicated that $\text{SO}_4^{\cdot-}$, $\cdot\text{OH}$, $\text{O}_2^{\cdot-}$ and $^1\text{O}_2$ all participated, with $\text{SO}_4^{\cdot-}$ and $^1\text{O}_2$ playing dominant roles. The catalyst retained over 80% degradation efficiency after five cycles.

External electric-field polarization can promote surface enrichment of Co and Ni in spent NCM and reconstruct the electronic structure, significantly increasing oxygen-vacancy content. The degradation rate of sulfamethoxazole with the polarized catalyst was enhanced to 1.61 times that of the untreated sample, with $\text{SO}_4^{\cdot-}$ and $^1\text{O}_2$ playing dominant roles [29]. Loading spent ternary cathode material onto a PTFE membrane to construct a catalytic membrane (LNCM-PTFE) achieved 96.4% removal of 2,4,6-trichlorophenol within 20 min at neutral pH, demonstrating the potential of coupled membrane catalysis technology. DFT calculations showed that the adsorption energy of defective LNCM with oxygen vacancies for PAA (-7.32 eV) is stronger than that of defect-free LNCM (-6.85 eV), indicating that oxygen vacancies promote the adsorption of peroxide molecules and cleavage of the O-O bond [30]. Chen et al. [31] integrated spent LIB cathode and anode materials to prepare $\text{CoO}@\text{C}$ catalyst, in which graphite acts as a stable support for CoO. This catalyst achieved complete degradation of atrazine within 15 min in PMS activation, with a rate constant of 0.185 min^{-1} . The $\text{CoO}@\text{C}/\text{PMS}$ system is dominated by $\cdot\text{OH}$ as the active species and shows good adaptability over a wide pH range (5.0–11.0). DFT calculations indicated that the adsorption energy of PMS on $\text{CoO}@\text{C}$ (-3.72 eV) is significantly higher than on pure carbon (-1.86 eV), and the O-O bond length of PMS in the $\text{CoO}@\text{C}/\text{PMS}$ system ($1.508\text{ \AA}$) is longer than that in the C/PMS system ($1.468\text{ \AA}$), demonstrating that CoO loading effectively promotes the activation and cleavage of the O-O bond in PMS. Zhou et al. [32] co-pyrolyzed spent LCO and biomass to prepare $\text{CoO}-\text{C}$ catalyst, which showed excellent performance in PMS-activated degradation of ciprofloxacin; the carbon coating effectively reduced Co leaching and improved electron distribution. It is worth noting that the Ni content in ternary materials has a marked effect on catalytic performance; high-Ni systems (e.g., NMC811) show better activity but relatively lower stability, while low-Ni systems (e.g., NMC111) show the opposite. This “activity-stability trade-off” is a key factor to be carefully considered in the design of spent ternary-material catalysts [12].

4.1.3. LiMn_2O_4 -Derived Catalysts: Direct Electrode Use and Manganese Retention

The catalytic activity of spinel-type LMO depends mainly on the $\text{Mn}^{3+}/\text{Mn}^{4+}$ redox couple. Wang et al. [6] directly used spent LMO electrode sheets as PMS catalysts for the degradation of o-phenylphenol (Figure 2c). They found that structural changes during battery cycling—such as surface reconstruction caused by Mn dissolution and re-deposition—can actually enhance catalytic performance. The catalyst maintained 94.8% degradation efficiency after 10 cycles, and the spent LMO electrode sheets could be used directly without complex pulverization, greatly simplifying the recovery process. Notably, the sulfate radicals generated during PMS activation can effectively suppress Mn dissolution, making LMO exhibit better stability in the PMS system than in pure water.

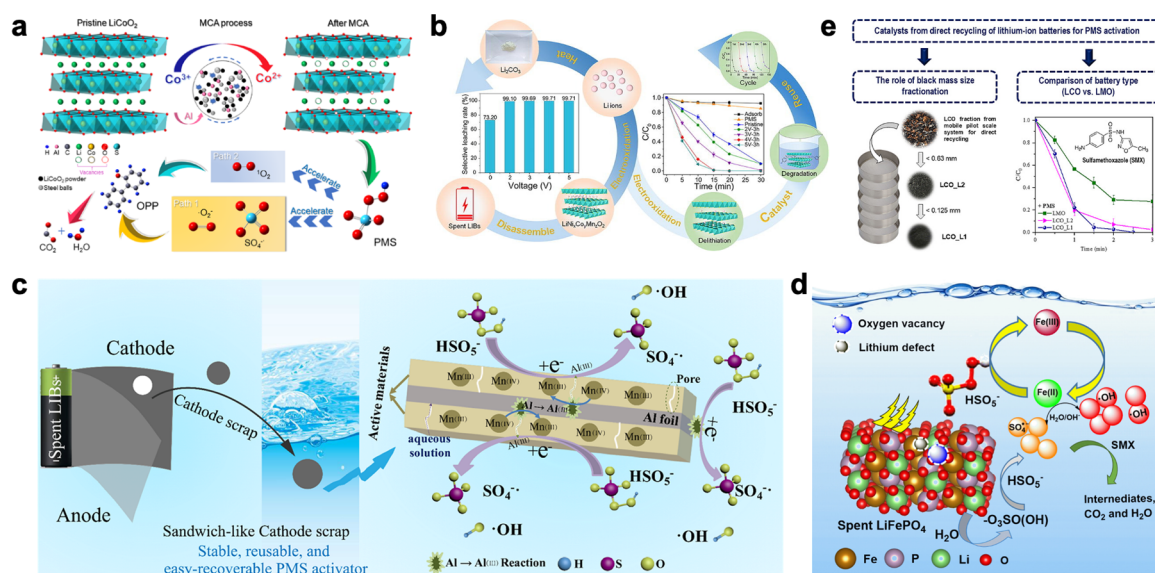


Figure 2. (a) Mechanochemical activation of spent LCO. Reproduced with permission. Ref. [7] Copyright 2021, American Chemical Society. (b) Electrochemical activation of spent NCM.; Reproduced with permission. Ref. [26] Copyright 2022, Elsevier. (c) Direct reuse of spent LMO cathodes. Reproduced with permission. Ref. [6] Copyright 2020, American Chemical Society. (d) Fenton-like activation by spent LFP. Reproduced with permission. Ref. [33] Copyright 2022, Elsevier. (e) Direct PMS activation by spent black mass. Reproduced with permission. Ref. [11] Copyright 2026, Elsevier.

4.1.4. LiFePO₄-Derived Catalysts: Iron-Centered Fenton-Like Activation

The Fe site in the olivine-structured LFP has a natural affinity for Fenton-like reactions. Wang et al. [33] found that, compared with well-ordered fresh LFP, retired LFP, rich in lithium defects and oxygen vacancies, exhibited stronger PMS activation capability, with a degradation rate for sulfamethoxazole much higher than that of the fresh-LFP-based control system (Figure 2d).

4.1.5. Spent Black Mass: Integrated Feedstocks and Compositional Heterogeneity

Kowalkińska et al. [11] directly used recovered spent black mass (mainly containing LCO and LMO) as a PMS catalyst; the smallest size fraction (LCO_L1) achieved rapid degradation of sulfamethoxazole within 2.5 min ($k_{app} = 1.815 \text{ min}^{-1}$) and efficiently removed 21 micropollutants from municipal wastewater effluent (Figure 2e). The reaction was dominated by a non-radical pathway ($^1\text{O}_2$). This study demonstrates that spent black mass can be used directly as a multifunctional catalyst without complex separation, providing a new approach for large-scale, high-value utilization of spent battery materials.

4.1.6. Cross-Chemistry Lessons for Oxidant Activation

Overall, the performance of spent cathode materials in PMS activation is distinctly material-dependent. LCO shows the most stable PMS activation performance because of its reversible $\text{Co}^{2+}/\text{Co}^{3+}$ cycle. NCM exhibits the most comprehensive behavior through electronic synergy among Ni, Co and Mn. LMO is limited by Mn dissolution, although sulfate radicals provide some stability by inhibiting this process. LFP, which relies on the Fenton affinity of Fe, offers distinct value in specific applications. In terms of reaction mechanisms, PMS activation by spent cathode materials usually involves both radical pathways ($\text{SO}_4^{\cdot-}$, $\cdot\text{OH}$) and non-radical pathways ($^1\text{O}_2$, surface-mediated electron transfer); their relative contributions depend on material composition, surface defect state and reaction conditions. Among these, $^1\text{O}_2$ has been confirmed as an important reactive species in many spent-cathode/PMS systems, and its generation pathways mainly include PMS self-decomposition, conversion of $\text{O}_2^{\cdot-}$, and oxygen-vacancy-mediated electron-transfer processes. Apparent pseudo-first-order rate constants reported across individual studies cannot be directly compared, because they are strongly sensitive to experimental variables including catalyst loading, PMS dosage, target pollutant identity, and pollutant concentration. To facilitate more meaningful cross-study evaluation of intrinsic catalytic performance, Table 1 summarizes key reaction parameters together with normalized kinetic metrics, including the surface-area-normalized rate constant and the PMS-concentration-normalized rate constant, for representative spent-cathode-

derived PMS activators. Here, k_{norm} is calculated as $k_{\text{norm}} = k/(\text{Cat. loading} \times \text{BET})$, where k is the apparent pseudo-first-order rate constant, catalyst loading is the catalyst dosage in g L^{-1} , and BET denotes the specific surface area ($\text{m}^2 \text{g}^{-1}$). These normalized descriptors partially offset differences in reaction conditions. Nevertheless, direct performance comparison remains valid only for identical target pollutants; material rankings derived from mixed-pollutant datasets should be interpreted with caution. Raw experimental conditions are retained alongside normalized values so readers can inspect the original test boundaries.

Table 1. Normalized performance comparison of representative spent cathode-derived PMS catalysts.

Catalyst	Pollutant	Conc. (mg L^{-1})	Cat. loading (g L^{-1})	PMS (mM)	k (min^{-1})	BET ($\text{m}^2 \text{g}^{-1}$)	k_{norm} ($\text{L min}^{-1} \text{m}^{-2}$)	$k/[\text{PMS}]$ ($\text{min}^{-1} \text{mM}^{-1}$)	Stability (Retention/Cycles)	Ref.
Spent LCO (CM-850)	LFX	10	0.4	0.5	0.076	1.47	0.129	0.152	80%/4 cycles	[5]
Spent NCM (NCM-650)	RhB	20	0.5	1	0.179	2.906	0.123	0.179	98.2%/4 cycles	[25]
CoFe ₂ O ₄ -LIBs	BPA	20	0.2	0.325	0.119	78.19	0.0076	0.366	50.5%/3 cycles	[27]
Black mass (LCO L1 fine)	SMX	10	1	0.5	1.815	6.5	0.279	3.63	~60–70% TOC/5 cycles	[11]
CoO@C (cathode + anode)	ATZ	5	0.05	0.5	0.185	26.553	0.139	0.37	>90%/5 cycles	[21]

Note: $k_{\text{norm}} = k/(\text{Cat. loading} \times \text{BET})$; k = apparent pseudo-first-order rate constant (min^{-1}); catalyst loading (g L^{-1}); BET = specific surface area ($\text{m}^2 \text{g}^{-1}$); $k/[\text{PMS}]$ denotes rate constant normalized by initial PMS molar concentration.

4.2. Graphite-Anode-Enabled H₂O₂ Electrosynthesis and Electro-Fenton Treatment

This subsection focuses on application-specific evidence for spent graphite in H₂O₂ electrosynthesis and electro-Fenton treatment. The studies below evaluate spent graphite as a cathode, an electro-Fenton interface and a conductive support for metal-based catalysts, complementing the material-attribute discussion in Section 2. Wei et al. [9] used nitric acid activation to treat graphite recovered from spent batteries, increasing oxygen-containing surface groups such as -COOH and C-O-C. The resulting catalyst achieved a Faradaic efficiency of approximately 80% for H₂O₂ electrosynthesis and removed approximately 92% of COD in high-COD wastewater, linking surface regulation to two-electron oxygen-reduction performance.

In electro-Fenton systems, the surface functional group composition of spent graphite directly determines its performance orientation. Cao et al. [8] systematically compared the influence of different recovery processes on the reuse of spent anode graphite in electro-Fenton. They found that acid leaching (AL) and combined acid-alkali leaching (AAL) treatments significantly changed the surface functional group composition of graphite powder, the AAL-treated graphite, having higher 2e⁻ ORR activity and H₂O₂ selectivity, outperformed the untreated powder and AL samples in H₂O₂ yield. However, when these graphite samples were used as electro-Fenton cathodes, the AL electrode exhibited the best degradation performance, achieving 100% removal of bisphenol A (BPA) within 70 min and 87.4% COD removal within 240 min. Mechanistic analysis indicated that the carboxylic acid content in AL powder was as high as 35.83% (far higher than 7% in AAL); these carboxyl groups can effectively adsorb ferric ions from solution, forming a heterogeneous iron redox system on the cathode surface, thereby promoting efficient Fenton reactions. More importantly, the AL cathode maintained 100% BPA removal after 10 cycles at low current density, demonstrating excellent reusability and environmental friendliness.

The high electrical conductivity of spent graphite also makes it an efficient support for supported catalysts. Chen et al. used graphite and copper recovered from spent LIB anodes, combined with iron oxides from steel-scale waste, to successfully prepare graphite-supported zero-valent iron-copper bimetallic catalyst (ZVI-Cu/C) via carbothermal reduction. By adjusting the mass ratio of steel-scale waste to spent anode powder (1:1 to 1:4), a series of catalysts were synthesized; among them, ZVI-Cu/C-2 showed the highest 4-chlorophenol removal in both reductive and heterogeneous Fenton reactions, with the degradation process following pseudo-first-order kinetics. The catalyst maintained high 4-CP removal activity after four cycles, with low leaching of iron and copper ions, confirming its good stability [17]. In addition, Guan et al. [34] prepared expanded-graphite-supported zero-valent iron (ZVI/EG) catalyst using spent LIB anode graphite and ferric chloride via carbothermal reduction; in the ZVI/EG/H₂O₂ heterogeneous Fenton system, 4-chlorophenol removal reached 97% within 1 h, with a degradation rate constant of 0.0527 min^{-1} , and still retained 82% removal after six cycles.

It is noteworthy that the study by Cao et al. [8] reveals a key design principle: the pretreatment strategy for spent graphite must match the target application scenario, for efficient H₂O₂ electrosynthesis, combined acid-alkali leaching should be chosen to enrich 2e⁻ ORR active sites; for rapid degradation of pollutants at the cathode surface, acid washing should be chosen to enrich carboxyl groups and promote surface Fenton cycling. This principle has important guiding significance for the design of spent-graphite-based catalytic materials.

4.3. Emerging Catalytic Pathways beyond Oxidant Activation and Electro-Fenton

Beyond the two main application categories above, the potential of spent-electrode-derived catalytic materials in water treatment extends to other directions, including photocatalytic degradation, reductive dechlorination, heavy-metal adsorption, and electrocatalytic nitrate reduction, but the relevant studies are relatively scattered, and their systematicity needs further strengthening.

In photocatalysis, Niu et al. [35] converted spent LiCoO_2 with melamine and NH_4Cl via one-pot sintering into Li-doped $\text{g-C}_3\text{N}_4/\text{Co}_3\text{O}_4$ composite photocatalyst; the photocatalytic H_2 production rate and Rhodamine B degradation rate were 8.7-fold and 6.8-fold higher than that of $\text{g-C}_3\text{N}_4$, respectively. Dhiman et al. [36] synthesized Co_3O_4 nanoparticles from spent LIBs, achieving 86% degradation of methyl blue within 45 min under sunlight. In reductive dechlorination, Chen et al. [17] used spent-graphite-supported ZVI-Cu/C bimetallic catalyst to simultaneously achieve reductive dechlorination and heterogeneous Fenton oxidation of 4-chlorophenol.

Overall, the application of spent-electrode-derived catalytic materials in water treatment has already covered persulfate activation, electro-Fenton, H_2O_2 electrosynthesis, photocatalysis and other directions, preliminarily forming a complete research chain from “material characterization-catalytic application-mechanistic elucidation”. Table 2 summarizes the representative studies of these systems, illustrating the compatibility of different spent electrode materials with specific catalytic processes. Among these, PMS activation is currently the most thoroughly studied and most mature system, with the redox cycling of transition metals in spent cathode materials showing the highest mechanistic compatibility with PMS activation; electro-Fenton, on the other hand, fully exploits the unique advantage of spent graphite as a conductive substrate, forming a complementary application pattern to the cathode-based approaches. However, most current studies are limited to batch experiments under ideal conditions; extension to real wastewater and continuous-flow reactors remains a critical challenge to be addressed.

5. Translational Challenges for Catalytic Upcycling

Promising pollutant-removal performance under laboratory conditions does not by itself establish the practical value of spent electrode-derived catalysts. Their catalytic properties depend on variable waste feedstocks, while their long-term use may introduce metal release, structural degradation, and performance losses in complex water matrices. Moreover, converting a waste material into a catalyst is not necessarily environmentally or economically preferable to metal recovery or direct electrode regeneration. The practical viability of catalytic upcycling therefore depends on four interconnected challenges: feedstock reproducibility, catalyst stability, realistic treatment performance, and system-level sustainability.

5.1. Feedstock Heterogeneity and Cross-Batch Reproducibility

Feedstock heterogeneity is an intrinsic feature of spent lithium-ion battery electrodes. At the chemistry level, commercial batteries contain layered LiCoO_2 and Ni-Mn-Co oxides, spinel oxides, LiFePO_4 , and various graphite-based anodes. Even within the same nominal chemistry, elemental ratios, particle coatings, conductive additives and binder formulations vary among manufacturers. These differences are further amplified by battery ageing, which depends on cycling depth, operating temperature, charging rate, storage history and failure mechanism. Consequently, spent electrodes with similar labels may exhibit substantially different lithium contents, defect densities, surface compositions and residual capacities. Recycling and pretreatment introduce another source of variability. Mechanical separation, thermal treatment, solvent washing, and acid leaching can alter surface functional groups, remove residual metals, or create new defects. Black mass may also contain mixed cathode chemistries, graphite, Cu, Al, binders, and electrolyte decomposition products. Some of these components act as catalytic promoters, whereas others block active sites or increase environmental risks. Therefore, bulk elemental composition alone is insufficient to predict catalytic behavior. The spatial distribution, chemical state, and accessibility of these components must also be considered. Current reproducibility tests commonly use repeated experiments from a single catalyst batch. Such technical replicates assess experimental precision but do not establish reproducibility across waste sources. Future studies should include independently collected electrode batches with different ageing histories and should report the distribution, rather than only the average, of catalytic performance.

Standard reference materials would further improve comparability. Pristine electrode materials, compositionally matched synthetic catalysts and representative black-mass mixtures could serve as complementary controls. Catalytic results should also be normalized using several descriptors, including catalyst mass, transition-metal content, accessible surface area and electrochemically active area. As illustrated in Table 1, such normalization reduces bias originating from divergent experimental set-ups across literature reports. These practices would help distinguish general structure-activity relationships from effects that are specific to a particular waste batch.

Table 2. Representative spent lithium-ion battery materials for advanced oxidation and electrochemical water treatment.

Catalytic System/Application	Key Material/Precursor	Reaction Conditions & Parameters	Performance Results	Catalytic Mechanism/Strategy	Reference
PMS Activation	Spent mobile LIB cathode (CM-850)	Catalyst 0.4 g L ⁻¹ , PMS 0.5 mM, pH = 4.65, LFX 10 mg L ⁻¹ , 60 min	LFX 94% (60 min); TOC 32%; Wide pH adaptability (3–9)	Calcination forms Co ²⁺ /Co ³⁺ valence cycling to activate PMS; synergy of radical (SO ₄ ^{•-}) and non-radical (¹ O ₂) pathways.	[5]
PMS Activation	Spent LiMn ₂ O ₄ cathode scrap	Catalyst = 1 piece scrap, Oxone = 0.3 g L ⁻¹ , OPP 20 mg L ⁻¹ , T = 25 °C, 60 min	OPP 94.8%; 94.8% after 10 cycles; Low metal leaching	Utilizes intrinsic “sandwich” structure and defects from lithiation/delithiation cycles to enhance activity.	[6]
PMS Activation	Spent LiCoO ₂ cathode + Al foil (MCA)	MCA activation, catalyst 0.2 g L ⁻¹ , PMS 0.2 g L ⁻¹ , pH = 7, OPP 20 mg L ⁻¹ , 30 min	OPP 100% (30 min); >96% after 5 cycles; Rate constant doubled	Al foil acts as in-situ reductant; MCA induces aluminothermic reduction, converting Co ³⁺ to Co ²⁺ and increasing oxygen vacancies.	[7]
PMS Activation	Spent LiCoO ₂ (SLCO-CM500)	Catalyst 0.2 g L ⁻¹ , PMS 2 mM, pH = 7, BAC 10 mg L ⁻¹ , 60 min	BAC 100% (60 min); TOC 90.4%; 1.9 × fresh Co ₃ O ₄	Utilizes intrinsic Li defects and oxygen vacancies (OVs) in spent cathode; electron transfer pathway (ETP) participates.	[19]
PMS Activation	Spent LiCoO ₂ scrap + DES	Catalyst = 1 piece scrap, Oxone = 0.2 g L ⁻¹ , pH≈7, OPP 20 mg L ⁻¹ , 60 min	OPP 98%; 95% after 10 cycles; Recovered 95% Co, 86% Li via DES	Structural disintegration after catalytic cycles promotes subsequent DES leaching for efficient Co/Li recovery (“catalysis & recovery” loop).	[10]
PMS Activation	Spent LIB cathode (CoFe ₂ O ₄ @CNTs)	Catalyst 35 mg L ⁻¹ , PMS 2.0 mM, pH = 6.5, CQP 10 mg L ⁻¹ , 25 min	CQP 98.7% (25 min); TOC 33%; >80% after 5 cycles	CNTs enhance electron transfer and inhibit Co leaching; SO ₄ ^{•-} , •OH, O ₂ ^{•-} , and ¹ O ₂ synergistically degrade.	[28]
PMS Activation	Spent LIB cathode & anode (CoO@C)	Catalyst 50 mg L ⁻¹ , PMS 0.5 mM, pH = 7, ATZ 5 mg L ⁻¹ , 15 min	ATZ 100% (15 min); >90% continuous flow	Synergistic preparation using Co source and defective graphite; DFT confirms strong PMS adsorption and charge transfer.	[31]
PMS Activation	Spent graphite (SNGO-2 N-doped graphene)	Catalyst 0.3 g L ⁻¹ , PMS 0.3 g L ⁻¹ , pH = 7, RhB 30 mg L ⁻¹ , 10 min	RhB 100% (10 min); TOC 83%; 4 cycles stable	Graphitic N is the main active center, achieving rapid degradation via non-radical pathway (¹ O ₂).	[37]
PMS Activation	Spent ternary cathode (NCM-650)	Catalyst 0.5 g L ⁻¹ , PMS 1 mM, pH = 4.85, RhB 20 mg L ⁻¹ , 25 min	RhB 100% (25 min); 98.2% after 4 cycles	Synergistic Co/Mn valence cycling (Co ²⁺ /Co ³⁺ , Mn ²⁺ /Mn ³⁺ /Mn ⁴⁺) activates PMS via radical and non-radical pathways.	[25]
PMS Activation	Spent LIB cathode (CoFe ₂ O ₄ -LIBs)	Catalyst 0.2 g L ⁻¹ , PMS 0.1 g L ⁻¹ , pH 7.6, BPA 20 mg L ⁻¹ , 25 min	BPA 94% (25 min); 1.7 × pure CoFe ₂ O ₄	Green leaching with citric acid/glucose; oxalic acid precipitation. High Co(II)/Co(III) ratio and more acid sites.	[27]
PMS Activation	Spent NCM (SG-NCM400)	Catalyst 0.2 g L ⁻¹ , PMS 0.7 mM, pH 5–9, MB 20 mg L ⁻¹ , 30 min (MB, SMX, LVF, RhB)	MB 90%; SMX 55%, LVF 79.5%, RhB 98.9%	Sol-gel reconstruction increases surface area to 85.6 m ² g ⁻¹ , increasing Co ²⁺ and Mn ²⁺ active sites.	[22]
PMS Activation	Spent LIB black mass (LCO_L1 fine)	Direct recycling, catalyst 1 g L ⁻¹ , PMS 0.5 mM, SMX 10 mg L ⁻¹ , 2.5 min	SMX 100% (2.5 min); k = 1.815 min ⁻¹ ; Degrades 21 micropollutants	Physical fractionation obtains Co-rich fine fraction; Low C/metal ratio dominates activation via non-radical ¹ O ₂ .	[11]
Electro-Fenton	Spent anode graphite (AL treatment)	1.25 mA cm ⁻² , pH = 3, Na ₂ SO ₄ 0.05 M, BPA 50 mg L ⁻¹ , 240 min	BPA 100% (70 min); COD 87.4% (240 min); Stable over 10 cycles	Acid leaching (AL) retains 35.83% -COOH groups, promoting Fe ion adsorption and accelerating interfacial Fe redox cycling.	[8]
Fenton and Fenton-like processes	Spent LiFePO ₄ (SULM)	Synergy with NH ₂ OH, pH = 3.5–3.8, catalyst 0.1 g L ⁻¹ , H ₂ O ₂ 4 mM	RhB 100% (30 min); TCH, AZY, ERY > 97%	NH ₂ OH promotes Fe(III)/Fe(II) cycling and synergy between interface and solution Fe; main active species are •OH and ¹ O ₂ .	[15]

Table 2. Cont.

Catalytic System/Application	Key Material/Precursor	Reaction Conditions & Parameters	Performance Results	Catalytic Mechanism/Strategy	Reference
Fenton and Fenton-like processes	Spent anode graphite + Mill Scale (ZVI-Cu/C)	Catalyst 0.2 g L ⁻¹ , H ₂ O ₂ 0.5 mM, pH = 3, 4-CP 10 mg L ⁻¹ , 120 min	4-CP 100% (120 min); 92.1% after 4 cycles	Carbothermal reduction forms Fe-Cu bimetallic synergy, promoting electron transfer.	[17]
Fenton and Fenton-like processes	Spent LiFePO ₄ (Iron hydroxyphosphate FPOH)	Hydrothermal conversion; pH 5–9, catalyst 1 g L ⁻¹ , H ₂ O ₂ 5 mL	Pb adsorption: 43.2 mg g ⁻¹ ; MB 100% (12 h)	Converts LiFePO ₄ to Fe ₅ (PO ₄) ₄ (OH) ₃ ·2H ₂ O; reacts with Pb to form stable lead hydroxyphosphate precipitation.	[38]
Fenton and Fenton-like processes	Spent graphite anode (AW+CT)	Catalyst 0.3 g L ⁻¹ , Fe ³⁺ 0.3 mM, H ₂ O ₂ 1 mM, pH = 3.0, BPA 30 min	k _{obs} = 0.1225 min ⁻¹ ; Li recovery 99.92%; LCA reduced by 20%	AW+CT removes impurities, retains surface Mn(II); Mn(II) promotes Fe(III)/Fe(II) cycling and ·OH generation.	[39]
Fenton and Fenton-like processes	Spent anode graphite (ZVI/EG)	ZVI/EG-2 0.4 g L ⁻¹ , H ₂ O ₂ 2.2 mM, pH = 3, 4-CP 10 mg L ⁻¹ , 60 min	4-CP 97% (60 min); 82% after 6 cycles; k = 0.0527 min ⁻¹	Chemical oxidation expands graphite as a support for ZVI, increasing surface area and Fe dispersion for ·OH production.	[34]
H ₂ O ₂ oxidation and electrosynthesis systems	Spent anode graphite (HNO ₃ activation)	RRDE test, 0.5 M H ₂ SO ₄ , Potential from 0.00 to −0.30 V (vs. SCE)	Faradaic efficiency 80–90%; Real T-acid wastewater COD removal > 92%	HNO ₃ introduces -COOH and C-O-C groups, activating adjacent C atoms for a highly selective 2e ⁻ ORR pathway.	[9]
H ₂ O ₂ oxidation and electrosynthesis systems	Spent LiCoO ₂ cathode tape (Li-BCT)	0.2 M H ₂ O ₂ ; MB 3 mg L ⁻¹ , 150 min (dark)	Rate constant increased 200×; Activation energy dropped from 83 to 26 kJ mol ⁻¹	Co ³⁺ /Co ²⁺ redox pair oxidizes H ₂ O ₂ to superoxide radicals (O ₂ ^{·-}), cleaving aromatic rings.	[40]
H ₂ O ₂ oxidation and electrosynthesis systems	Spent mobile LIB cathode (LiCoO ₂)	CCRD optimization: H ₂ O ₂ 1.71 M, MB 6 mg L ⁻¹ , LiCoO ₂ 32 mg, T = 32 °C, 90 min	MB 93.3%	Multivariate optimization (RSM); Co ²⁺ catalyzes ·OH generation, but excess H ₂ O ₂ causes quenching.	[41]
Other catalytic systems and anode valorization	Spent LIB cathode (CoFe ₂ O ₄ -LIBs)	pH = 3, catalyst 30 mg, H ₂ O ₂ 8 mL (3%), MB 4.8 mg L ⁻¹ , 420 min	MB 87.7% (420 min); Comparable to commercial CoFe ₂ O ₄	Recovered Co used to synthesize spinel nano-ferrite; synergistic Co ²⁺ /Co ³⁺ and Fe ²⁺ /Fe ³⁺ catalysis generates ·OH.	[42]
Other catalytic water-treatment systems	Spent LiCoO ₂ + Melamine + NH ₄ Cl	Simulated sunlight; RhB degradation	RhB degradation rate: 6.8 × higher than g-C ₃ N ₄	Li-doped g-C ₃ N ₄ /Co ₃ O ₄ heterojunction; Li narrows bandgap, Co ₃ O ₄ builds a built-in electric field for charge separation.	[35]
Other catalytic water-treatment systems	Spent LiCoO ₂ + Melamine + NH ₄ Cl (LiClCo-C ₃ N ₄)	Simulated sunlight; RhB degradation	RhB degradation rate: 15.3 × higher than g-C ₃ N ₄	Li-Cl-Co regulates π-conjugation; Li/Cl intercalation alters spacing, reducing charge transport barriers.	[43]
Other catalytic systems and anode valorization	Spent LNCM (LNCM-PTFE)	Membrane filtration, PAA 0.3 mM, pH = 7.0, TCP 2 mg L ⁻¹ , 20 min	TCP 96.4% (20 min); High real wastewater efficiency	Oxygen vacancies (OVs) promote charge transfer and PAA cleavage; main species: carbon-centered radicals and ¹ O ₂ .	[30]
Other catalytic systems and anode valorization	Spent LIB cathode (Co/Co ₃ O ₄)	Electrodeposited Co + 0.27 M H ₂ O ₂ + UV; Azo dye 0.25 g L ⁻¹ , 210 min	Azo dye 85% (210 min); Rate constant 12 × H ₂ O ₂ alone	Co is oxidized in-situ to spinel Co ₃ O ₄ by H ₂ O ₂ ; UV excitation generates holes (h ⁺) and electrons (e ⁻).	[44]

5.2. Metal Release, Active-Phase Evolution and Operational Durability

Pollutant loss in a nominally heterogeneous system should not automatically be assigned to a surface pathway. Co, Ni, Mn, Fe and Cu may be released through proton-promoted dissolution, ligand-assisted complexation, oxidant-induced corrosion or surface reconstruction. Dissolved ions can then activate PMS, PDS, H₂O₂ or PAA homogeneously, whereas metal-containing colloids may pass ordinary filtration and behave differently from either molecular ions or the parent solid. Operationally, a surface-mediated contribution requires activity that remains associated with the retained catalyst; a homogeneous contribution is supported when the catalyst-free filtrate continues consuming oxidant or removing pollutant at a rate reproduced by dissolved-metal controls prepared at the measured concentrations.

Mechanistic discrimination should combine time-resolved ICP-MS or ICP-OES measurements with hot-filtration tests, matched dissolved-metal controls, and ultrafiltration, dialysis or membrane-separation experiments that distinguish ions from colloidal particles. Sampling should cover the initial burst-release period and subsequent steady operation, while pH, ligands, oxidant concentration and ionic strength are held constant. The recovered solid should be re-tested and characterized by XPS, XRD and electron microscopy to determine whether activity loss or retention follows metal depletion, valence changes or formation of a new surface phase. Chelating agents can provide supporting evidence, but they should not be used alone because they may also alter oxidant chemistry and surface adsorption.

A complete balance for each metal should account for its initial inventory and its distribution among the residual catalyst, dissolved fraction, colloidal or nanoparticulate fraction, reactor-wall deposits, recovered catalyst and intentionally recovered metal stream. Studies should report balance closure, cumulative release as a percentage of the initial metal inventory, and release normalized to catalyst mass, treated-water volume and pollutant mass mineralized. Existing studies illustrate the reporting gap: cleaned LiMn₂O₄ retained 94.8% pollutant removal after 10 cycles but described leaching only qualitatively as low [6], whereas acid-leached graphite maintained complete BPA removal over 10 cycles [8]. More recently, AW+CT-treated spent graphite was evaluated in continuous flow and reported 99.92% Li recovery together with a total-metal-ion safety metric of 4.1341 mg L⁻¹ [39]. These endpoints are useful, but they still do not replace a closed balance that partitions dissolved, colloidal and deposited metal fractions.

5.3. Performance Resilience in Realistic Water Matrices

The transition from purified water to real wastewater changes both reaction kinetics and dominant treatment mechanisms. Laboratory studies commonly use one model pollutant at relatively high concentration in buffered or ultrapure water. Under these conditions, most generated reactive species are available for target-pollutant degradation. Real water matrices contain natural organic matter, inorganic ions, suspended particles and mixtures of contaminants that compete for reactive species and catalyst surfaces. Bicarbonate, carbonate, chloride, phosphate and natural organic matter can scavenge radicals, coordinate dissolved metals or modify oxidant activation pathways. Organic matter and suspended solids may also block active sites or foul porous electrodes. Variations in pH, conductivity and dissolved oxygen can alter electrochemical reactions and H₂O₂ production. These effects are catalyst- and mechanism-dependent. A matrix component that inhibits radical oxidation may have a smaller influence on a selective non-radical pathway, whereas chloride may produce reactive chlorine species and change both degradation efficiency and product toxicity.

Real-water validation should therefore follow a staged testing framework. Initial experiments can use defined model solutions to identify intrinsic reaction mechanisms. Synthetic matrices containing controlled concentrations of major ions and organic matter can then isolate individual interference effects. The final stage should use actual municipal or industrial wastewater, preferably without relying exclusively on artificially spiked target compounds. Paired tests using the same catalyst in model and real matrices would allow the magnitude and origin of performance losses to be quantified. Treatment performance should not be assessed solely through disappearance of the parent pollutant. Relevant endpoints include total organic carbon, COD, oxidant utilization, transformation-product formation and residual toxicity. For electrochemical systems, energy consumption, current efficiency and electrode fouling should also be reported. Long-term flow-through operation is needed to evaluate hydraulic behaviour, catalyst attrition and the accumulation of matrix-derived deposits. Seasonal and batch-to-batch variations in wastewater composition should ultimately be incorporated because a single real-water sample cannot represent operational variability.

5.4. Life-Cycle Environmental and Techno-Economic Feasibility

A quantitative sustainability comparison requires a common functional unit and system boundary. For catalytic upcycling, the primary functional unit should be 1 m³ of wastewater treated to a defined removal,

mineralization and toxicity endpoint, supplemented by 1 kg of spent electrode processed. The boundary should include battery collection, discharge and dismantling; electrode separation; solvent, acid, alkali and oxidant production; washing, drying and heating; catalyst-conversion yield; reactor electricity and oxidant demand; catalyst recovery and lifetime; cumulative metal release; regeneration; and final metal recovery or disposal. Reporting only catalyst-synthesis energy, or only pollutant-removal efficiency, can reverse the apparent ranking when catalyst lifetime and treatment-stage inputs are included.

The available quantitative evidence is encouraging but heterogeneous. Directly cleaned LiMn_2O_4 retained 94.8% OPP removal after 10 cycles, although preparation energy and a closed metal balance were not reported [6]. For mild activation, acid-leached spent graphite maintained complete BPA removal over 10 cycles [8]. An acid-washing-plus-calcination route for spent graphite reported 99.92% Li recovery, an electrical energy per order of $402.44 \text{ kWh m}^{-3} \text{ order}^{-1}$, a total-metal-ion safety metric of 4.1341 mg L^{-1} and a 20% reduction in environmental burden relative to commercial graphite [39]. In another short-process example, LiCoO_2 recovered through discharging, dismantling, alkaline washing and calcination was used for PAA activation; its LCA reported reductions of 67% in carbon emissions, 92% in human toxicity and 75% in cost relative to purchasing LiCoO_2 [45]. These comparisons support catalytic reuse against virgin catalyst supply, but they do not yet establish superiority to conventional battery recycling.

Conventional recycling LCAs provide useful contextual benchmarks. Ciez and Whitacre compared pyrometallurgical, hydrometallurgical and direct cathode recycling and found that direct cathode recovery can retain more embodied value, with reported break-even refunctionalization costs of approximately $\text{US\$15 kg}^{-1}$ for NMC and $\text{US\$19 kg}^{-1}$ for NCA [3]. For NMC111 hydrometallurgy, Iturrondobeitia et al. reported global-warming potentials of $25.1\text{--}95.2 \text{ kg CO}_2 \text{ eq kg}^{-1}$ recycled cathode. Most evaluated routes consumed approximately $1\text{--}5 \text{ kWh kg}^{-1}$ cathode, whereas bioleaching required 14 kWh kg^{-1} ; reagent demand ranged from 0.78 kg kg^{-1} cathode for bioleaching to 42.75 kg kg^{-1} for a lactic-acid/ H_2O_2 route [46]. These data show that a reagent described as green does not necessarily produce a lower life-cycle burden. To our knowledge, no direct head-to-head LCA has yet compared direct reuse, mild activation and deep reconstruction as water-treatment catalysts with conventional metal recovery using the same feedstock, treatment endpoint and functional unit. Table 3 therefore presents reported values as evidence benchmarks rather than as a normalized ranking.

Table 3. Quantitative evidence for the process intensity and sustainability of catalytic upcycling routes.

Route	Conversion Intensity and Inputs	Catalyst Lifetime and Metal-Release Evidence	Reported LCA/TEA Evidence	Ref.
Direct reuse	Discharge, dismantling, washing and/or physical size fractionation; no intentional phase reconstruction. Preparation energy and reagent inventories are generally not reported.	Cleaned LiMn_2O_4 retained 94.8% OPP removal after 10 cycles, but metal leaching was reported only qualitatively as low [6].	—	[6]
		—	No cradle-to-grave LCA was found for direct reuse as a water-treatment catalyst. Conventional direct cathode recycling provides only a contextual benchmark [3].	[3]
Mild activation	Acid/alkali washing, electrochemical treatment or thermal activation up to approximately 500°C ; fewer unit operations than complete dissolution and resynthesis, but preparation energy and reagent mass are seldom reported.	Acid-leached graphite maintained complete BPA removal for 10 cycles [8].	—	[8]
		For acid-washing-plus-calcination route graphite, Li recovery was 99.92%, the reported total-metal-ion safety metric was 4.1341 mg L^{-1} , and continuous-flow stability was assessed [39].	Acid-washing-plus-calcination route graphite reduced environmental burdens by 20% relative to commercial graphite [39]. A four-step recovered LiCoO_2 /PAA route reported 67% lower carbon emissions, 92% lower human toxicity and 75% lower cost than purchasing LiCoO_2 [45].	[39] [45]
Deep reconstruction	Complete or extensive leaching, separation, precipitation/sol-gel reconstruction, washing, drying and calcination. A conventional NMC111 hydrometallurgical benchmark required approximately $1\text{--}5 \text{ kWh kg}^{-1}$ cathode for most routes and 14 kWh kg^{-1} for bioleaching.	—	For conventional NMC111 hydrometallurgy, reported GWP ranged from 25.1 to $95.2 \text{ kg CO}_2 \text{ kg}^{-1}$ cathode and reagent inputs from 0.78 to 42.75 kg kg^{-1} cathode [46].	[46]

6. Future Prospects—Research Priorities for Deployable Catalytic Upcycling

6.1. Developing Simple and Low-Energy Conversion Methods

The environmental value of converting spent lithium-ion battery electrodes into catalysts depends not only on catalytic activity, but also on whether the conversion process consumes fewer chemicals and less energy than conventional metal recovery and catalyst synthesis. Many current approaches involve repeated acid leaching, precipitation, washing, drying, and high-temperature calcination. Although these treatments can produce well-defined catalytic phases, they may weaken the environmental advantages of using waste-derived feedstocks. Future research should therefore adopt a process-intensity hierarchy in which direct reuse, mild surface activation, and partial phase reconstruction are prioritized over complete dissolution and resynthesis. For spent graphite anodes, simple washing, low-concentration acid treatment, electrochemical activation, or controlled oxidation may be sufficient to remove unstable surface residues while retaining conductive graphite frameworks. For cathode materials, selective delithiation may offer a more efficient route than complete metal separation, as it can expose redox-active transition-metal sites while preserving the multimetallic framework. Electrochemical delithiation, mechanochemical activation, microwave heating, and flash Joule heating are particularly promising because they shorten reaction times and can induce defects, phase reconstruction, or elemental redistribution without prolonged furnace treatment. However, a short treatment time does not necessarily indicate a low-energy process. Future studies should report the energy and material inputs associated with electrode separation, reagent production, washing, drying, off-gas treatment, and catalyst recovery. Combining process-level energy analysis with techno-economic and life-cycle assessments will therefore be necessary to determine whether catalytic upcycling yields a genuine environmental benefit. The overall research framework for catalytic upcycling of spent lithium-ion battery electrodes is illustrated in Figure 3.

6.2. Stabilization Strategies for Safe Catalyst Operation

The transition metals responsible for the catalytic activity of spent cathode materials, particularly Co, Ni, and Mn, can also become secondary contaminants if they are released during water treatment. Catalyst stability should therefore be treated as a central performance criterion rather than an additional characterization item. Apparent pollutant degradation may partially originate from dissolved metal ions, especially under acidic conditions or during persulfate activation. Future studies should distinguish heterogeneous surface reactions from homogeneous reactions by combining metal-leaching measurements, filtration experiments, dissolved-metal controls, and post-reaction structural characterization. Material design should balance accessible active sites with structural stability. Possible strategies include anchoring cathode-derived metal species onto recovered graphite, constructing self-supported electrodes, producing magnetically recoverable composites, and introducing thin ion-permeable protective layers. Residual Cu and Al from current collectors should not automatically be classified as unwanted impurities because they may modify electron transfer, active-site dispersion, or interfacial redox cycling. Nevertheless, beneficial effects must be evaluated together with the potential release of Cu, Co, Ni, Mn, Li, F, and particulate carbon. Stability tests should extend beyond several repeated batch cycles. More meaningful protocols would quantify activity retention, metal release, and structural evolution over prolonged operation under different pH values and in the presence of chloride, bicarbonate, phosphate, and natural organic matter. Metal release should be normalized to both the volume of water treated and the amount of pollutant removed. In addition, parent-compound disappearance should be accompanied by measurements of mineralization, transformation products and residual toxicity. The eventual regeneration, metal recovery, or safe disposal of the deactivated catalyst must also be incorporated into the proposed process. Such an end-of-second-life strategy is essential to prevent catalytic upcycling from creating a new stream of metal-containing waste.

6.3. Real-Wastewater Validation and Continuous-Flow Reactor Engineering

Most spent-electrode-derived catalysts have been evaluated in batch reactors containing a single pollutant in ultrapure or buffered water. These systems are useful for identifying reaction pathways, but they provide limited information about practical treatment performance. Real wastewaters contain mixtures of organic contaminants, inorganic ions, suspended solids and natural organic matter that can compete for reactive species, block active sites or alter metal speciation. Future studies should therefore progress from single-compound solutions to pollutant mixtures, simulated complex matrices and, ultimately, untreated or biologically treated industrial and municipal effluents. Removal efficiency should be complemented by COD, total organic carbon, acute toxicity and targeted transformation-product analysis. Continuous-flow systems are particularly important because they reveal limitations associated with mass transfer, catalyst attrition, pressure drop, electrode polarization and long-

term fouling. Spent graphite can potentially be fabricated into gas-diffusion cathodes, porous flow-through electrodes, or packed catalytic columns, whereas cathode-derived oxides can be immobilized on conductive supports, membranes, or monolithic carriers. More broadly, flow-through electro-Fenton studies using conventional carbon electrodes have shown that electrode arrangement, flow direction, and residence-time distribution strongly influence H_2O_2 utilization and mineralization in real wastewater. These reactor principles can guide the engineering of spent-electrode-based systems. A staged validation framework is recommended. Laboratory studies should initially determine intrinsic kinetics and matrix effects. Bench-scale flow reactors should then evaluate hydraulic behavior, catalyst retention and energy consumption. Pilot-scale studies should finally assess fluctuating wastewater composition, maintenance requirements and integration with biological or membrane treatment. Relevant performance indicators include space-time yield, electrical energy per order, oxidant utilization efficiency, pressure drop, metal release per cubic meter of treated water, and catalyst lifetime. Reporting these parameters would enable meaningful comparisons with established advanced oxidation and electrochemical treatment technologies.

6.4. AI-Enabled Feedstock-to-Reactor Optimization

Artificial intelligence and machine learning could help address a defining challenge of spent-battery-derived catalysts: the strong variability of the starting materials. Electrode composition, state of health, degradation mechanism, residual electrolyte, and current-collector contamination vary between manufacturers and battery histories. Consequently, an identical conversion procedure may generate catalysts with substantially different structures and activities. Machine-learning models could connect feedstock characteristics with conversion conditions, catalyst properties and water-treatment performance, thereby supporting feedstock classification and route selection. A useful database should record battery chemistry and provenance, elemental composition, phase structure, surface chemistry, defect density, conversion conditions and water-matrix composition. Model outputs should extend beyond pollutant removal and include mineralization, reaction selectivity, metal leaching, catalyst lifetime, energy consumption and cost. Multi-objective models could then identify conversion and operating conditions that balance activity, stability, environmental safety, and process intensity. These studies provide transferable concepts, although equivalent frameworks have not yet been systematically validated for catalysts derived from spent battery electrodes. Because available datasets are likely to remain small and heterogeneous, future work should avoid relying exclusively on complex models. Physics-informed machine learning, transfer learning, Bayesian optimization, and active learning may be more appropriate because they can incorporate electrochemical and catalytic knowledge while reducing the number of experiments required. Model uncertainty should be explicitly reported, and validation should separate battery chemistries, electrode batches, and wastewater types between training and testing datasets. Ultimately, AI should be integrated into a closed-loop workflow. Rapid characterization and online water-quality sensors would supply inputs to predictive models. The models would recommend conversion or reactor conditions, and experimental outcomes would then be used to update them. Such a framework could link waste-electrode sorting, catalyst preparation, and reactor operation within a single data-driven platform. However, its success will depend on standardized reporting, open and machine-readable datasets, and mechanistic constraints that prevent statistically accurate but chemically unrealistic predictions.

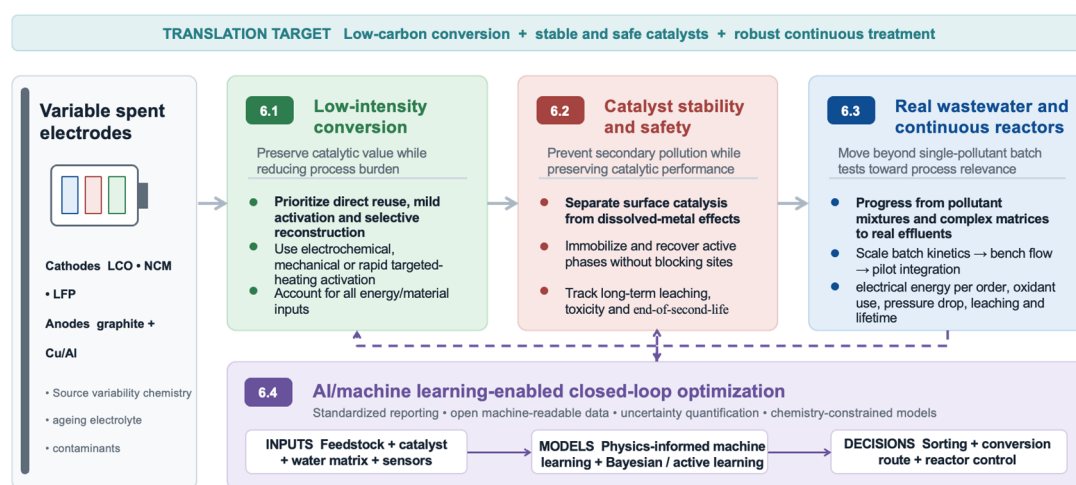


Figure 3. Future directions for catalytic upcycling of spent LIB electrodes.

Author Contributions

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Use of AI and AI-Assisted Technologies

During the preparation of this work, the manuscript language was polished by the authors with the assistance of DeepSeek, an AI language model. DeepSeek was used solely for language polishing and correction of grammar errors. After using this service, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

Abbreviations

2e [−] ORR	two-electron oxygen reduction reaction
4-CP	4-chlorophenol
AAL	acid-alkali-leached
AI	artificial intelligence
AL	acid-leached
ATZ	atrazine
AZY	azithromycin
BAC	benzalkonium chloride
BET	Brunauer-Emmett-Teller
BPA	bisphenol A
ChCl	choline chloride
CNTs	carbon nanotubes
COD	chemical oxygen demand
CQP	chloroquine phosphate
DFT	density functional theory
DES	deep eutectic solvent
EG	expanded graphite
EPR	electron paramagnetic resonance
ERY	erythromycin
ETP	electron transfer pathway
GWP	global warming potential
H ₂ O ₂	hydrogen peroxide
ICP-MS	inductively coupled plasma mass spectrometry
ICP-OES	inductively coupled plasma optical emission spectrometry
LCA	life cycle assessment
LCO	lithium cobalt oxide
LFP	lithium iron phosphate
LFMP	lithium iron manganese phosphate
LFX	levofloxacin

LIB	lithium-ion battery
Li-ion	lithium-ion
LNCM	lithium nickel cobalt manganese oxide
LMO	lithium manganese oxide
LVF	levofloxacin
MB	methylene blue
MCA	mechanochemical activation
NCA	lithium nickel cobalt aluminum oxide
NCM	lithium nickel cobalt manganese oxide
NMC	lithium nickel manganese cobalt oxide
NMC111	$\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$
NMC811	$\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$
NMP	N-methyl-2-pyrrolidone
O_2	oxygen
$\text{O}_2^{\cdot-}$	superoxide radical
OER	oxygen evolution reaction
OPP	o-phenylphenol
ORR	oxygen reduction reaction
PAA	peracetic acid
PDS	peroxydisulfate / persulfate
PMS	peroxymonosulfate
PTFE	polytetrafluoroethylene
PVDF	polyvinylidene fluoride
RhB	rhodamine B
RP	raw powder
SG	sol-gel
SLCO	spent LiCoO_2
SMX	sulfamethoxazole
SNGO	spent graphite-derived N-doped graphene
$\text{SO}_4^{\cdot-}$	sulfate radical
SULM	sea-urchin-like material
TCH	tetracycline hydrochloride
TEA	techno-economic assessment
TOC	total organic carbon
US\$	United States dollar
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffraction
ZVI	zero-valent iron
ZVI-Cu/C	graphite/carbon-supported zero-valent iron-copper bimetallic catalyst
ZVI/EG	expanded-graphite-supported zero-valent iron
$\cdot\text{OH}$	hydroxyl radical
k_{app}	apparent pseudo-first-order rate constant
k_{norm}	normalized rate constant
$k/[\text{PMS}]$	rate constant normalized by initial PMS molar concentration
Cat. loading	catalyst loading
Conc.	concentration
vol%	volume percent
Oxone	commercial peroxymonosulfate
Li^+	lithium ion
e^-	electron
Fe^0	zero-valent iron
$\text{Fe}^{3+}/\text{Fe}^{2+}/\text{Fe}^0$	iron redox states
$\text{Co}^{2+}/\text{Co}^{3+}$	cobalt redox states
$\text{Mn}^{2+}/\text{Mn}^{3+}/\text{Mn}^{4+}$	manganese redox states
$\text{Fe(II)}/\text{Fe(III)}$	iron oxidation states
$\text{Co(II)}/\text{Co(III)}$	cobalt oxidation states
P^{3-}	phosphide anion
O-O	peroxide bond
Li-O	lithium-oxygen bond
CM-850	spent mobile LIB cathode catalyst label
LCO_L1	black mass
LNCM-PTFE	LNCM-loaded PTFE catalytic membrane
NCM-650	calcined spent NCM catalyst label
SG-NCM400	sol-gel-derived NCM catalyst label
SLCO-CM500	spent LiCoO_2 catalyst label
SNGO-2	sample 2 of SNGO
ZVI-Cu/C-2	sample 2 of ZVI-Cu/C
$\text{CoFe}_2\text{O}_4@\text{CNTs}$	cobalt ferrite supported on carbon nanotubes
$\text{CoO}@\text{C}$	cobalt oxide encapsulated in carbon
CoO-C	cobalt oxide-carbon composite

References

1. Harper, G.; Sommerville, R.; Kendrick, E.; et al. Recycling lithium-ion batteries from electric vehicles. *Nature* **2019**, *575*, 75–86. <https://doi.org/10.1038/s41586-019-1682-5>.
2. Schwarzenbach, R.P.; Escher, B.I.; Fenner, K.; et al. The challenge of micropollutants in aquatic systems. *Science* **2006**, *313*, 1072–1077. <https://doi.org/10.1126/science.1127291>.
3. Ciez, R.E.; Whitacre, J.F. Examining different recycling processes for lithium-ion batteries. *Nat. Sustain.* **2019**, *2*, 148–156. <https://doi.org/10.1038/s41893-019-0222-5>.
4. Shi, R.; Wang, B.; Tang, D.; et al. Towards high value-added recycling of spent lithium-ion batteries for catalysis application. *Electrochem. Energy Rev.* **2024**, *7*, 28. <https://doi.org/10.1007/s41918-024-00220-1>.
5. Zhao, Y.; Yuan, X.; Jiang, L.; et al. Reutilization of cathode material from spent batteries as a heterogeneous catalyst to remove antibiotics in wastewater via peroxymonosulfate activation. *Chem. Eng. J.* **2020**, *400*, 125903. <https://doi.org/10.1016/j.cej.2020.125903>.
6. Wang, X.; Zhang, X.; Dai, L.; et al. Recycling the cathode scrap of spent lithium-ion batteries as an easily recoverable peroxymonosulfate catalyst with enhanced catalytic performance. *ACS Sustain. Chem. Eng.* **2020**, *8*, 11337–11347. <https://doi.org/10.1021/acssuschemeng.0c03389>.
7. Dang, S.; Zhou, P.; Shi, P.; et al. *In Situ* Aluminothermic Reduction Induced by Mechanochemical Activation Enhances the Ability of the Spent LiCoO₂ Cathode to Activate Peroxymonosulfate. *ACS Sustain. Chem. Eng.* **2021**, *9*, 15375–15385. <https://doi.org/10.1021/acssuschemeng.1c03583>.
8. Cao, Z.; Zheng, X.; Cao, H.; et al. Efficient reuse of anode scrap from lithium-ion batteries as cathode for pollutant degradation in electro-Fenton process: Role of different recovery processes. *Chem. Eng. J.* **2018**, *337*, 256–264. <https://doi.org/10.1016/j.cej.2017.12.104>.
9. Wei, J.; Wang, X.; Wu, X. Recycle graphite from spent lithium-ion batteries for H₂O₂ electrosynthesis. *Environ. Sci. Pollut. Res.* **2023**, *30*, 98183–98194. <https://doi.org/10.1007/s11356-023-29354-3>.
10. Guo, H.; Min, Z.; Hao, Y.; et al. Sustainable recycling of LiCoO₂ cathode scrap on the basis of successive peroxymonosulfate activation and recovery of valuable metals. *Sci. Total Environ.* **2021**, *759*, 143478. <https://doi.org/10.1016/j.scitotenv.2020.143478>.
11. Kowalkińska, M.; Hupka, Ł.; Ostrowski, A.; et al. Valorization of size-fractionated black mass from spent lithium-ion batteries for catalytic peroxymonosulfate activation. *Sustain. Mater. Technol.* **2026**, *48*, e02085. <https://doi.org/10.1016/j.susmat.2026.e02085>.
12. Brillas, E.; Martínez-Huitle, C.A. Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. An updated review. *Appl. Catal. B Environ.* **2015**, *166–167*, 603–643. <https://doi.org/10.1016/j.apcatb.2014.11.016>.
13. Wang, M.; Lei, L.; Zhao, C.; et al. From Spent Batteries to Green Hydrogen: Catalytic Upcycling of Lithium-Ion Battery Cathodes for Water Electrolysis. *Adv. Mater.* **2026**, *38*, e23322. <https://doi.org/10.1002/adma.202523322>.
14. Chen, M.; Zhou, Y.; Wang, L.; et al. Recycling spent LiNi_{1-x-y}Co_xMn_yO₂ cathodes to efficient catalysts for the oxygen evolution reaction. *Green Chem.* **2024**, *26*, 2912–2921. <https://doi.org/10.1039/d3gc04503j>.
15. Zou, W.; Li, J.; Wang, R.; et al. Hydroxylamine mediated Fenton-like interfacial reaction dynamics on sea urchin-like catalyst derived from spent LiFePO₄ battery. *J. Hazard. Mater.* **2022**, *431*, 128590. <https://doi.org/10.1016/j.jhazmat.2022.128590>.
16. Dhanabalan, K.; Sriram, G.; Oh, T.H. An Overview of Electrocatalysts Derived from Recycled Lithium-Ion Batteries for Metal–Air Batteries: A Review. *Energies* **2025**, *18*, 4933. <https://doi.org/10.3390/en18184933>.
17. Chen, S.; Long, F.; Gao, G.; et al. Zero-valent iron-copper bimetallic catalyst supported on graphite from spent lithium-ion battery anodes and mill scale waste for the degradation of 4-chlorophenol in aqueous phase. *Sep. Purif. Technol.* **2022**, *286*, 120466. <https://doi.org/10.1016/j.seppur.2022.120466>.
18. Feng, N.; Wang, J.; Lin, Y.; et al. Recycling Electrode Materials of Spent Lithium-Ion Batteries for High-Efficiency Catalyst Application: Recent Advances and Perspectives. *Energy Fuels* **2024**, *38*, 19174–19187. <https://doi.org/10.1021/acs.energyfuels.4c02887>.
19. Lin, S.; Niu, B.; Shi, X.; et al. Non-destructive approach for upcycling the cathode of spent lithium-ion batteries: Combined with the efficient treatment of organic wastewater. *Sep. Purif. Technol.* **2025**, *360*, 130917. <https://doi.org/10.1016/j.seppur.2024.130917>.
20. Liu, X.; Ni, J.; Yin, J.; et al. Turning Waste into Treasure: Directly Utilizing Ternary Lithium-Ion Battery Cathode Waste for an Efficient Oxygen Evolution Reaction through Defect Engineering. *Energy Fuels* **2025**, *39*, 18043–18052. <https://doi.org/10.1021/acs.energyfuels.5c02782>.
21. Ji, G.; Tang, D.; Wang, J.; et al. Sustainable upcycling of mixed spent cathodes to a high-voltage polyanionic cathode material. *Nat. Commun.* **2024**, *15*, 4086. <https://doi.org/10.1038/s41467-024-48181-9>.

22. Xu, L.; Liang, L.; Chen, C.; et al. Upcycling spent lithium battery cathodes into efficient PMS catalysts for organic contaminants degradation. *J. Environ. Chem. Eng.* **2023**, *11*, 111605. <https://doi.org/10.1016/j.jece.2023.111605>.
23. Kotkar, A.; Dash, S.; Bhanja, P.; et al. Microwave assisted recycling of spent Li-ion battery electrode material into efficient oxygen evolution reaction catalyst. *Electrochim. Acta* **2023**, *442*, 141842. <https://doi.org/10.1016/j.electacta.2023.141842>.
24. Wang, Y.; Zhou, Y.; Chen, M.; et al. Recycling spent lithium-ion battery cathodes to multimetallic phosphides for high-efficiency oxygen evolution reaction. *ACS Sustain. Chem. Eng.* **2025**, *13*, 2477–2486. <https://doi.org/10.1021/acssuschemeng.4c08960>.
25. Zhao, Y.; Wang, H.; Ji, J.; et al. Recycling of waste power lithium-ion batteries to prepare nickel/cobalt/manganese-containing catalysts with inter-valence cobalt/manganese synergistic effect for peroxymonosulfate activation. *J. Colloid Interface Sci.* **2022**, *626*, 564–580. <https://doi.org/10.1016/j.jcis.2022.06.112>.
26. Dang, S.; Hou, W.; Min, Y.; et al. Electro-oxidation: A win-win strategy for the selective recovery of Li⁺ from spent lithium-ion batteries and the preparation of highly active catalysts. *Chem. Eng. J.* **2022**, *435*, 135169. <https://doi.org/10.1016/j.cej.2022.135169>.
27. Liang, J.; Xue, Y.; Gu, J.N.; et al. Sustainably recycling spent lithium-ion batteries to prepare magnetically separable cobalt ferrite for catalytic degradation of bisphenol A via peroxymonosulfate activation. *J. Hazard. Mater.* **2022**, *427*, 127910. <https://doi.org/10.1016/j.jhazmat.2021.127910>.
28. Hu, Z.; Luo, J.; Xu, S.; et al. Activation of peroxymonosulfate using spent Li-ion batteries for the efficient degradation of chloroquine phosphate. *Catalysts* **2023**, *13*, 661. <https://doi.org/10.3390/catal13040661>.
29. Zhou, D.; Tang, R.; Min, Y.; et al. External electric field-assisted electronic restructuring of transition metal oxides derived from spent lithium-ion batteries to enhance persulfate activation. *Appl. Surf. Sci.* **2023**, *625*, 157120. <https://doi.org/10.1016/j.apsusc.2023.157120>.
30. Feng, L.; Liao, X.; Jiang, F.; et al. Oxygen vacancies-enriched spent lithium-ion battery cathode materials loaded catalytic membrane for effective peracetic acid activation and organic pollutants degradation. *J. Hazard. Mater.* **2024**, *480*, 136444. <https://doi.org/10.1016/j.jhazmat.2024.136444>.
31. Chen, H.; Wei, Y.; Han, C.; et al. Peroxymonosulfate activation by CoO@C catalyst from integrated recycling of spent lithium-ion battery: Performance and mechanism. *Sep. Purif. Technol.* **2025**, *355*, 129658. <https://doi.org/10.1016/j.seppur.2024.129658>.
32. Zhou, F.; Liu, M.; Li, X.; et al. Preparation of CoO-C catalysts from spent lithium-ion batteries and waste biomass for efficient degradation of ciprofloxacin via peroxymonosulfate activation. *Chem. Eng. J.* **2023**, *471*, 144469. <https://doi.org/10.1016/j.cej.2023.144469>.
33. Wang, P.; Lou, X.; Chen, Q.; et al. Spent LiFePO₄: An old but vigorous peroxymonosulfate activator for degradation of organic pollutants in water. *Environ. Res.* **2022**, *214*, 113780. <https://doi.org/10.1016/j.envres.2022.113780>.
34. Guan, J.; Li, Z.; Chen, S.; et al. Zero-valent iron supported on expanded graphite from spent lithium-ion battery anodes and ferric chloride for the degradation of 4-chlorophenol in water. *Chemosphere* **2022**, *290*, 133381. <https://doi.org/10.1016/j.chemosphere.2021.133381>.
35. Niu, B.; Xiao, J.; Xu, Z. Recycling Spent LiCoO₂ Battery as a High-efficient Lithium-doped Graphitic Carbon Nitride/Co₃O₄ Composite Photocatalyst and Its Synergistic Photocatalytic Mechanism. *Energy Environ. Mater.* **2023**, *6*, e12312. <https://doi.org/10.1002/eem2.12312>.
36. Dhiman, S.; Gupta, B. Co₃O₄ nanoparticles synthesized from waste Li-ion batteries as photocatalyst for degradation of methyl blue dye. *Env. Technol. Innov.* **2021**, *23*, 101765. <https://doi.org/10.1016/j.eti.2021.101765>.
37. Yan, S.; Chen, X.; Yang, Y.; et al. Peroxymonosulfate activation by N-doped 3D graphene from spent lithium-ion batteries for organic pollutants degradation: An insight into the degradation mechanism. *Chem. Eng. J.* **2024**, *484*, 149379. <https://doi.org/10.1016/j.cej.2024.149379>.
38. Xu, L.; Chen, C.; Huo, J.B.; et al. Iron hydroxyphosphate composites derived from waste lithium-ion batteries for lead adsorption and Fenton-like catalytic degradation of methylene blue. *Environ. Technol. Innov.* **2019**, *16*, 100504. <https://doi.org/10.1016/j.eti.2019.100504>.
39. Peng, Y.; Zhang, Q.; Fan, Y.; et al. Recycling of spent graphite anodes in Fenton-like catalysis for green economy and long-term sustainable wastewater purification. *Chem. Eng. J.* **2025**, *520*, 165825. <https://doi.org/10.1016/j.cej.2025.165825>.
40. Abreu Gonçalves, M.C.; Garcia, E.M.; Taroco, H.A.; et al. Chemical recycling of cell phone Li-ion batteries: Application in environmental remediation. *Waste Manag.* **2015**, *40*, 144–150. <https://doi.org/10.1016/j.wasman.2015.02.014>.
41. de Matos Vargas, A.M.; da Silva Santos, C.; Cagliari, J.V.; et al. Multivariate optimization of the degradation conditions of methylene blue using a catalyst recovered from electronic waste. *J. Environ. Chem. Eng.* **2019**, *7*, 103191. <https://doi.org/10.1016/j.jece.2019.103191>.
42. Moura, M.N.; Barrada, R.V.; Almeida, J.R.; et al. Synthesis, characterization and photocatalytic properties of nanostructured CoFe₂O₄ recycled from spent Li-ion batteries. *Chemosphere* **2017**, *182*, 339–347. <https://doi.org/10.1016/j.chemosphere.2017.05.036>.

43. Niu, B.; Xiao, J.; Xu, Z. Utilizing spent Li-ion batteries to regulate the π -conjugated structure of g-C₃N₄: A win-win approach for waste recycling and highly active photocatalyst construction. *J. Mater. Chem. A* **2021**, 9, 472–481. <https://doi.org/10.1039/d0ta10881b>.
44. Falqueto, J.B.; Magnago, L.B.; Rocha, A.K.S.; et al. Photocatalytic properties of Co/Co₃O₄ films recycled from spent Li-ion batteries. *Ionics* **2018**, 24, 2167–2173. <https://doi.org/10.1007/s11581-018-2614-3>.
45. Li, G.; Cheng, Y.; Wu, Y.; et al. Recycling spent lithium-ion batteries as high-performance catalysts for peracetic acid activation toward rapid degradation of organic pollutants. *Chem. Eng. J.* **2026**, 538, 176664. <https://doi.org/10.1016/j.cej.2026.176664>.
46. Iturrondobeitia, M.; Vallejo, C.; Berroci, M.; et al. Environmental Impact Assessment of LiNi_{1/3}Mn_{1/3}Co_{1/3}O₂ Hydrometallurgical Cathode Recycling from Spent Lithium-Ion Batteries. *ACS Sustain. Chem. Eng.* **2022**, 10, 9798–9810. <https://doi.org/10.1021/acssuschemeng.2c01496>.