Chemical Science

ROYAL SOCIETY OF CHEMISTRY

PERSPECTIVE

View Article Online
View Journal

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

Check for updates

Cite this: DOI: 10.1039/d6sc02626e

All publication charges for this article have been paid for by the Royal Society of Chemistry

Recycling nickel-rich cathodes toward structural and functional circularity: a perspective

Yuhan Huang,[a] Haoyang Xu,[a] Yimin Zeng,[b] Han Xue*[b] and Ge Li*[a]

Received 31st March 2026
Accepted 12th May 2026

DOI: 10.1039/d6sc02626e

rsc.li/chemical-science

The increasing use of nickel-rich (Ni ≥ 0.6) layered oxide cathodes in lithium-ion batteries creates an urgent need for recycling, for which existing elemental-recovery methods are not well suited. Unlike process-based classifications, the present framework evaluates recycling routes by whether they recover only the elemental inventory, preserve the layered crystal framework, or restore and intentionally improve electrochemical function. Here, we reconceptualize cathode recycling as three progressive levels of material circularity: elemental recovery, structural preservation, and functional restoration. The specific degradation modes of Ni-rich cathodes, including surface rock–salt reconstruction, cation disorder, and intergranular fracture, not only limit the effectiveness of conventional pyrometallurgical and hydrometallurgical processes but also define the precise materials science targets that higher-level recycling strategies must address. Against this framework, we critically assess advances in direct recycling and upcycling, highlighting relithiation chemistry, molten-salt recrystallization, and Artificial Intelligence-guided process control as the most capable approaches for achieving structural and functional circularity at scale. Closing the loop for Ni-rich cathodes requires more than just incremental improvements to existing recovery methods. It calls for a strategic shift in recycling research to preserve and restore the cathode's crystal structure.

1 Introduction

The rapid adoption of electric vehicles (EVs) and renewable energy technologies has driven unprecedented demand for high-energy lithium-ion batteries (LIBs).[1] Among available cathode chemistries, Ni-rich layered oxides have emerged as the dominant choice for long-range EV applications. These materials are typically classified as layered transition-metal oxides, with a nickel mole fraction in the transition-metal layer of at least 0.6. Commercially significant examples include Nickel Manganese Cobalt Oxide (NMC), such as $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ (NMC 622), $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (NMC 811), $LiNi_{0.9}Co_{0.1}O_2$ (NMC 90). Nickel Cobalt Aluminum Oxide (NCA) is another important class of layered oxides that generally feature high nickel content, though specific formulations may vary.[2] The importance of this threshold is discussed in Section 2. Below a nickel mole fraction of 0.6, structural and thermal instabilities that would arise at higher nickel contents are mostly suppressed by residual Co and Mn. In contrast, above this threshold, these failure modes dominate the performance limitations.[3] Consequently, increasing the nickel content in this chemistry leads to exceptional gravimetric energy density, rising from approximately 160 mAh g^{-1} for NMC 111 to around 200 mAh g^{-1} for NMC 811, but at the cost of structural fragility, with direct and often underappreciated implications for end-of-life recyclability.[4–6]

As depicted in Fig. 1, the widespread use of Ni-rich cathodes enhances performance but also poses sustainability challenges, underscoring the need for closed-loop recycling. The features that make these materials excellent for energy storage also make their end-of-life management more challenging. This perspective concerns Co-containing Ni-rich compositions (NMC 622, NMC 811, NCA, and related materials) specifically. Although Co-free, high-Ni cathodes such as $LiNiO_2$-based materials are an active area of development,[6] their recycling challenges differ in important ways from those of Co-bearing compositions and lie outside the scope of the present discussion. Therefore, this perspective emphasizes the coupling between cathode chemistry and recyclability: recycling strategies for Ni-rich cathodes should aim to retain material value rather than simply convert structurally complex electrodes into elemental feedstocks.

1.1 From cobalt to nickel: composition, performance, and the origins of structural fragility

Understanding why Ni-rich cathodes present a distinct recycling challenge requires tracing their compositional evolution from earlier $LiCoO_2$ (LCO)-dominated chemistries. In LCO, Co stabilizes the layered $R\bar{3}m$ crystal structure through strong Co–O bonding, which resists phase transformation, and provides

---

[a] Department of Mechanical Engineering, University of Alberta, Edmonton, AB, Canada. E-mail: ge.li@ualberta.ca

[b] Natural Resources Canada, CanmetMATERIALS, Hamilton, Canada

© 2026 The Author(s). Published by the Royal Society of Chemistry

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

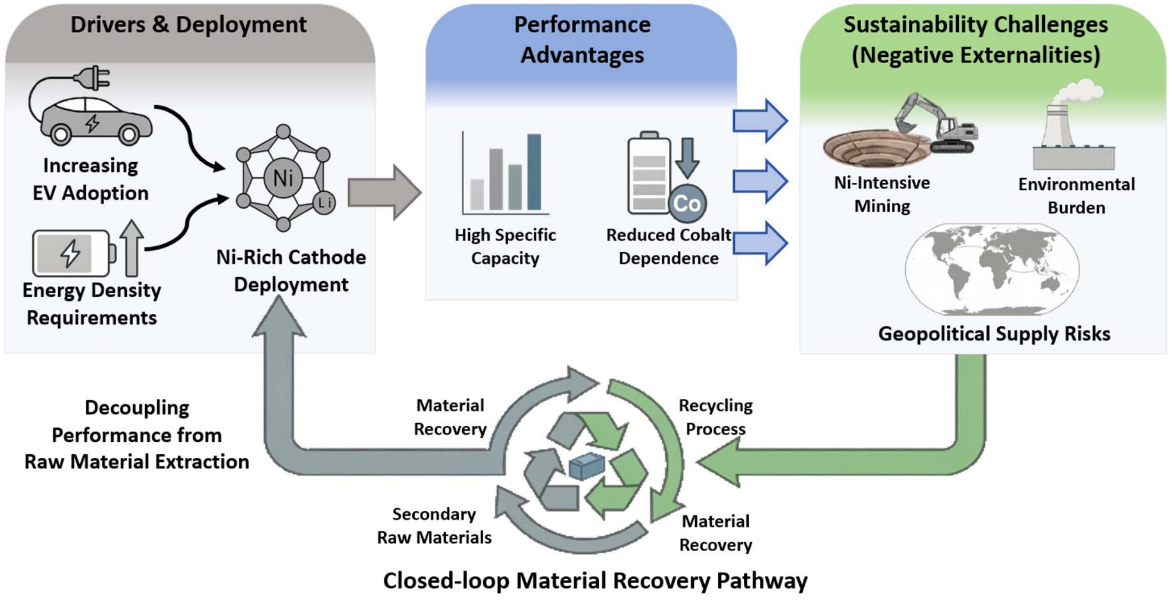

Fig. 1 System-level rationale for closed-loop recycling of Ni-rich cathodes. The rapid growth of EV deployment is increasing the volume of end-of-life LIBs, while the shift toward Ni-rich cathodes raises energy density and lowers cobalt intensity but also amplifies structural and interfacial degradation. Together, these trends create a need for closed-loop recycling routes that recover not only critical elements, but, where possible, the structural and functional value embedded in the spent cathode.

high electronic conductivity within the transition-metal layer, enabling fast charge/discharge kinetics. Despite these structural merits, Co carries severe drawbacks. It is costly, geopolitically concentrated (over 70% of global supply originates from the Democratic Republic of Congo), and raises serious toxicity and ethical sourcing concerns.[7–9] These pressures drove the battery industry toward partial Co substitution with Ni and Mn.

In $LiNi_xMn_yCo_zO_2$ (NMC; x, y, z are mole fractions of Ni, Mn, Co; $x + y + z = 1$) compounds, Ni^{2+}/Ni^{+} redox couples deliver the majority of usable capacity, Mn^{4+} acts as a structurally inactive electrochemical pillar that suppresses phase transformations, and Co^{3+} primarily maintains electronic conductivity and orderly Li^+ extraction kinetics.[6] Increasing the Ni fraction from NMC 111 ($x = 0.33$) to NMC 532 ($x = 0.50$), NMC 622 ($x = 0.60$), and NMC 811 ($x = 0.80$) generally increases practical capacity. At the same time, it reduces cobalt content and lowers the cost per unit energy.[4,5,10] The parallel NCA family achieves similarly high capacity through Al^{3+} doping, which reinforces structural integrity in the absence of Mn. As shown in Fig. 2a, the trajectory toward higher Ni fractions in the $LiCoO_2$–$LiNiO_2$–$LiMnO_2$ ternary space consistently moves compositions into the high-capacity but reduced-safety region.[11] Fig. 2b quantifies the underlying trade-off. Across the NMC series, practical capacity rises from approximately 160 to over 200 mAh g^{-1} as Ni content increases, while thermal stability drops from above 300 °C to below 230 °C, and capacity retention deteriorates markedly.[11] The nickel content that maximizes energy density also increases the severity and spatial heterogeneity of degradation during cycling. This connection is especially important for recycling, but it remains underexplored. As illustrated in Fig. 2c, spent NCM811 accumulates three compounding structural deficits, namely particle cracking, lithium loss with Li/Ni cation mixing, and surface rock–salt impurity formation, each of which must be addressed by any credible recycling strategy.[12] Fig. 2d contrasts the responses of regeneration and upcycling to these deficits: while regeneration restores the layered framework through lithium supplementation alone, upcycling introduces structural modifications that produce a closed-loop output exceeding the performance of the original material.[12]

1.2 A framework for cathode circularity: three levels of material recovery

The field of Ni-rich cathode recycling has, until recently, been organized almost entirely around elemental recovery, which means extracting nickel, cobalt, manganese, and lithium as metal salts to re-enter the supply chain. Existing LIB recycling categorizations are commonly process-centered, distinguishing pyrometallurgy, hydrometallurgy, and direct recycling/regeneration by their unit operations or product forms. By contrast, the framework proposed here is value-centered: it classifies recycling routes by the degree of electrochemical value retained in the spent cathode, from elemental recovery to structural preservation to functional restoration. Here, functional restoration is defined as the recovery and deliberate optimization of electrochemical performance, such as capacity, rate capability, cycle life, and interfacial stability. Therefore, the recycled cathode matches or exceeds the performance level of the original material, rather than merely preserving its elemental composition or crystal structure. We propose that a more productive organizing framework distinguishes three ascending levels of material circularity, as illustrated in Fig. 3. Material circularity is therefore used here differently from conventional recycling-efficiency metrics. Recovery yield,

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

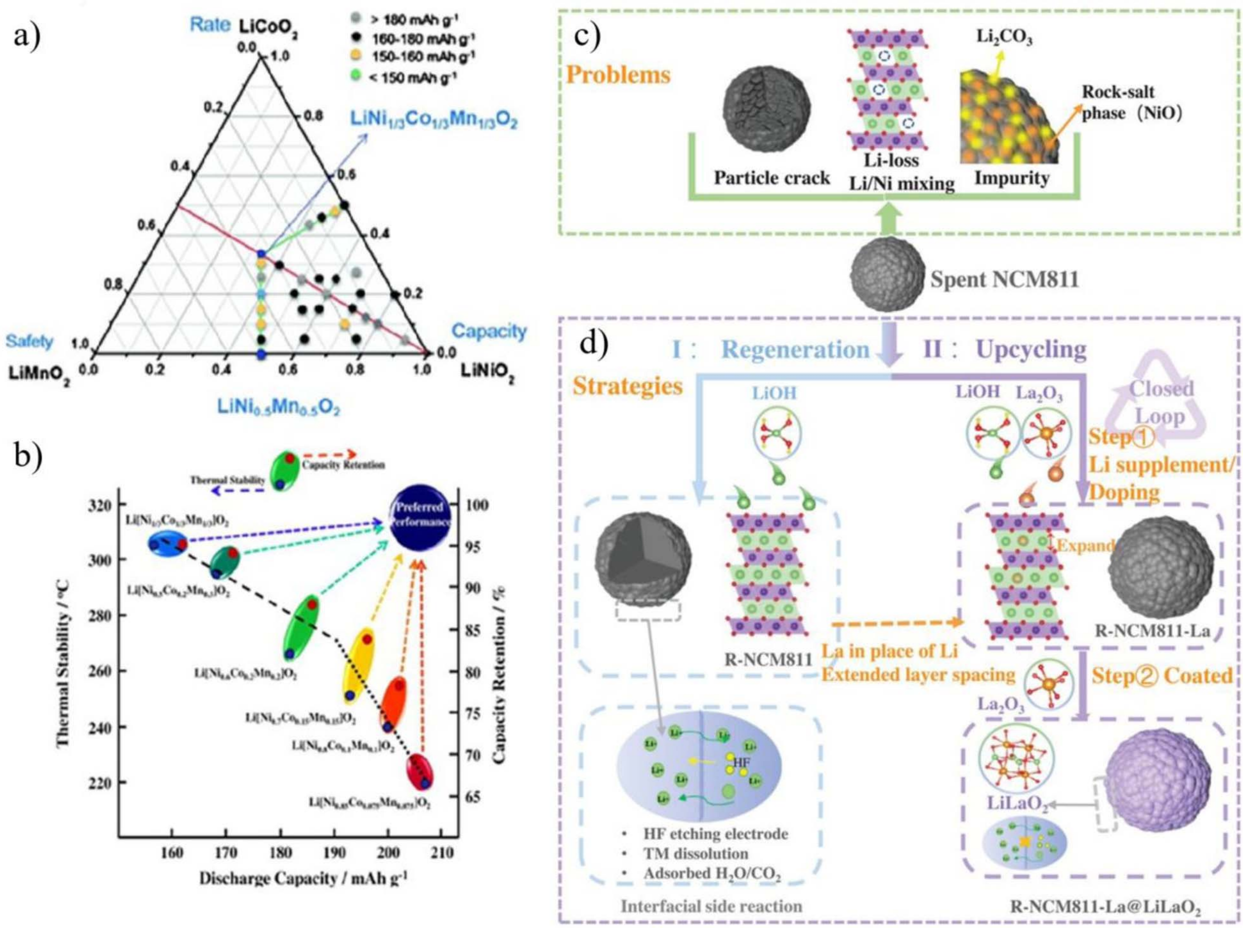

Fig. 2 (a) Ternary phase diagram of the $LiCoO_2$–$LiNiO_2$–$LiMnO_2$ system, showing that progression toward higher Ni fractions increases discharge capacity while narrowing safety margins. (b) Quantitative trade-off between discharge capacity, thermal stability, and capacity retention across the NMC series: as Ni content rises, capacity exceeds 200 mAh g^{-1}, but thermal stability drops below 230 °C, rendering spent Ni-rich cathodes both more valuable and more structurally compromised than lower-Ni counterparts. Panels (a) and (b) reproduced with permission from ref. 6. Copyright 2024 Korean Chemical Society. Compositional landscape of Ni-rich cathodes and the motivation for structural recycling. (c) The three principal degradation modes in spent NCM811: particle cracking, lithium loss with Li/Ni cation mixing, and surface rock–salt phase formation, which collectively define the material-science targets for Level II and Level III recycling. (d) Comparison of direct regeneration (Level II) and upcycling (Level III) strategies applied to spent NCM811, illustrating how lithium supplementation alone restores the layered framework, while additional La doping and $LiLaO_2$ surface coating yield a closed-loop product exceeding the performance of the original material. Panels (c) and (d) reproduced with permission from ref. 12. Copyright 2024 Wiley-VCH.

product purity, reagent consumption, energy input, and cost describe how efficiently a process recovers material mass or elemental value. Material circularity, by contrast, asks what level of electrochemically valuable information is retained or restored in the recycled cathode: only the elemental inventory, the layered crystal framework, or the battery function itself. In this sense, two processes with similar metal recovery efficiencies may represent very different levels of circularity if one destroys the cathode lattice, whereas the other preserves or upgrades it.

Level I (elemental recovery) includes pyrometallurgy and traditional hydrometallurgy. These methods dissolve or burn away the cathode structure completely, thereby retrieving metals in ionic or elemental form. The cathode crystal structure, which holds the material's electrochemical value, is permanently destroyed. Level II (structural preservation) involves direct recycling methods aimed at maintaining or restoring the $R\bar{3}m$ layered structure of the used cathode, replenishing lithium, and fixing cation disorder without full breakdown. Level III (functional restoration, or upcycling) refers to recycling routes in which the spent cathode is not only structurally repaired but intentionally engineered so that its electrochemical function matches or exceeds that of the original material. This functional improvement may be achieved through strategies such as single-crystal conversion, dopant introduction, compositional retuning, or surface architecture redesign. In this sense, functional restoration is distinguished from Level II structural preservation by the deliberate optimization of the recycled product's future battery performance, rather than by lattice restoration alone. Fig. 3 should therefore be read not as a conventional process-flow comparison, but as a value-retention map across three distinct recovery objectives: recovering elements, preserving structure, and restoring or upgrading function.

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

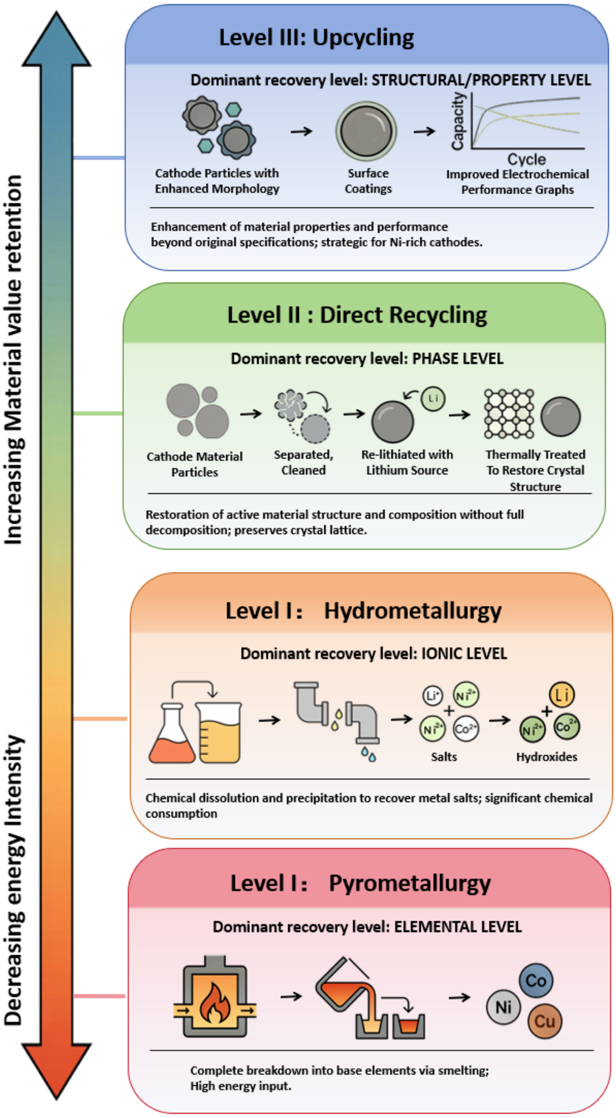

Fig. 3 Value-retention map of lithium-ion battery recycling strategies, comparing conventional elemental recovery with higher-level structural preservation and functional restoration in terms of recovery objective, energy intensity, and retained electrochemical value.

This framework is used throughout to evaluate existing recycling methods (Section 2), assess the state of emerging technologies (Section 3), and identify the scientific bottlenecks that must be resolved to make Level II and Level III recycling economically viable at scale (Section 5). Accordingly, the central question is not only whether critical metals can be recovered, but which degradation features must be repaired if higher-value circularity is to be credible. Section 2, therefore, maps the dominant failure modes of Ni-rich cathodes—surface rock–salt reconstruction, cation disorder, and intergranular fracture—onto the material-repair targets required for Levels II and III.

1.3 Why Ni-rich cathodes specifically demand a structural recycling approach

Within the broader field of nickel-containing cathode materials, the case for prioritizing structural and functional recycling hinges on three main arguments that are less applicable to lower-Ni compositions. First, Ni-rich cathodes currently dominate EV cell production and will constitute the vast majority of the end-of-life stream reaching recyclers by 2030.[13] Recycling infrastructure and process chemistry must therefore be optimized specifically for high-Ni chemistry. Second, the elevated nickel content in spent Ni-rich cathodes represents a far greater per-tonne recoverable value than lower-Ni compositions. Selectively recovering and directly reusing structurally intact high-Ni active material, rather than dissolving it into generic metal salts, unlocks economic returns that are unavailable for LCO or low-Ni NMC.[14] Third, the specific degradation modes of Ni-rich cathodes, namely surface rock–salt reconstruction, intergranular cracking, and cation disorder, are substantially more severe than those observed in lower-Ni counterparts.[4,15] These degradation modes are not just barriers to recycling, but also the specific advanced recycling targets that Level II and Level III strategies must address, as discussed in the following section.

2 Degradation mechanisms and the limits of elemental recovery

2.1 The three principal degradation modes and their recycling implications

2.1.1 Surface reconstruction and rock–salt phase formation. As schematically summarized in Fig. 4, degradation in Ni-rich cathodes is a progressive, multiscale process in which chemical, structural, and mechanical failure mechanisms evolve concurrently over repeated electrochemical cycling.

Lattice expansion and contraction during cycling drive lithium loss and transition-metal migration at the cathode particle surface, triggering the reconstruction of the electrochemically active $R\bar{3}m$ layered phase into a rock–salt-type ($Fm\bar{3}m$) surface layer with poor Li^+ conductivity.[16,17] As shown in Fig. 5c, this transformation proceeds *via* rate-dependent pathways. Low cycling rates favor extensive Li vacancy accumulation and disordered rock–salt formation, whereas high cycling rates drive an intermediate spinel phase.[15] The progressive nature of this reconstruction is directly visualized in Fig. 5d, where atomic-resolution High-Angle Annular Dark-Field Scanning Transmission Electron Microscopy (HAADF-STEM) imaging of NMC76 confirms that the $Fm\bar{3}m$ phase dominates the near-surface region after 200 cycles while bulk $R\bar{3}m$ ordering is retained.[18] Fig. 5a further demonstrates the competing layered-to-spinel transition at higher cycling rates, with fast Fourier transform (FFT) analysis confirming $Fd\bar{3}m$ spinel formation and transition-metal occupancy of Li layer positions after 200 cycles.[18]

This rock–salt layer thickens progressively with cycling and cannot be reversed by any Level I recovery process. Pyrometallurgy destroys it through smelting, and hydrometallurgy dissolves it along with the bulk material, but neither process restores the layered structure in the recovered product. Surface impurities, including LiOH and Li_2CO_3, further accelerate degradation by promoting parasitic reactions and CO_2

© 2026 The Author(s). Published by the Royal Society of Chemistry

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

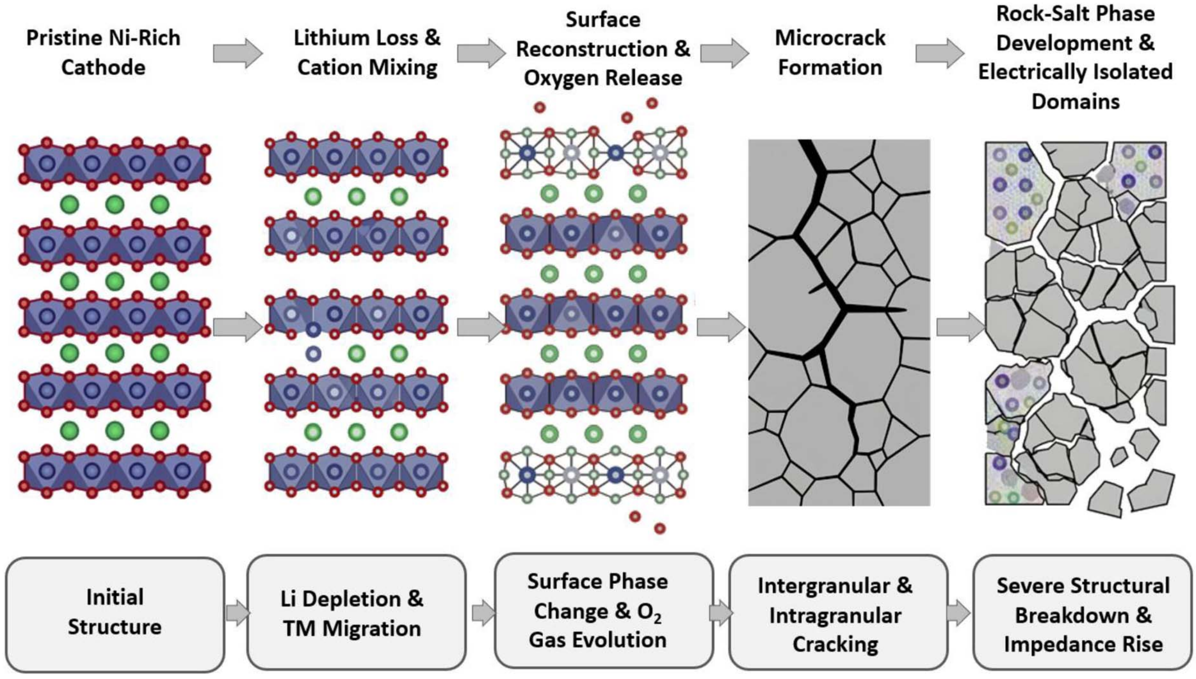

Fig. 4 Multiscale degradation mechanisms of Ni-rich cathode materials during electrochemical cycling.

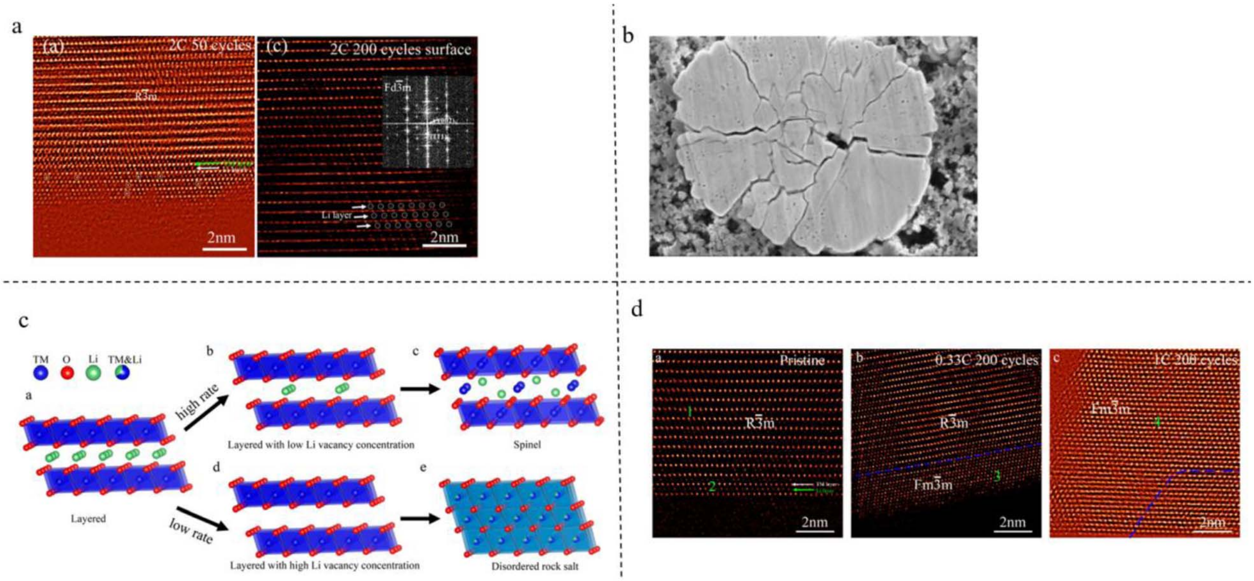

Fig. 5 The three principal degradation modes of Ni-rich cathodes and their implications for recycling strategy. (a) HAADF-STEM images of the competing layered-to-spinel transition at 2C, with FFT analysis confirming $Fd\bar{3}m$ spinel formation and transition-metal occupancy of Li sites after 200 cycles. (b) Focused ion beam-scanning electron microscopy (FIB-SEM) cross-section of a cycled NMC secondary particle showing intergranular crack networks that create electrically isolated domains and accelerate capacity decay. Together, these three mechanisms, surface phase reconstruction, cation disorder, and mechanical fracture, define the structural targets that Level II and III recycling strategies must address. (c) Schematic of rate-dependent phase transformation pathways from the pristine $R\bar{3}m$ structure: high cycling rates favour spinel formation, while low rates drive disordered rock–salt phase development. (d) HAADF-STEM images of NMC76 showing progressive surface rock–salt reconstruction with increasing cycle number, from intact $R\bar{3}m$ in the pristine state to a dominant $Fm\bar{3}m$ surface layer after 200 cycles. Panels (a, b and d) reproduced with permission from ref. 15. Copyright 2023 Royal Society of Chemistry. Panel (c) reproduced with permission from ref. 14. Copyright 2020 MDPI.

evolution.[3] This mechanism defines the primary target for Level II direct recycling. Any process that claims to restore electrochemical performance must demonstrate that it can dissolve or convert the rock–salt surface layer and re-establish a coherent $R\bar{3}m$ surface termination.

2.1.2 **Cation disorder and lithium deficiency.** Progressive lithium depletion during cycling is accompanied by the

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

migration of transition-metal cations, particularly Ni^{2+}, whose ionic radius closely matches that of Li^+, into lithium sites (*i.e.*, 3b Wyckoff positions).[19] The resulting cation disorder blocks Li^+ diffusion channels and reduces ionic conductivity. Recent *operando* neutron diffraction and four-dimensional scanning transmission electron microscopy studies further show that Li/Ni disordering contributes to heterogeneous structural evolution within Ni-rich cathodes, linking local defect distributions to non-uniform delithiation and degradation.[20] This disorder is invisible to Level I recovery processes: a hydrometallurgical product containing the correct stoichiometric ratio of nickel, cobalt, manganese, and lithium does not necessarily reconstitute into an ordered $R\bar{3}m$ phase upon re-synthesis unless processing conditions are carefully controlled.[21] Cation disorder, therefore, imposes a composition-control requirement on Level II strategies that is more stringent than the simple stoichiometric recovery that Level I achieves.[22]

2.1.3 Intergranular and intragranular fracture. The repeated anisotropic volume change of Ni-rich cathode particles during cycling generates mechanical stress that propagates as intergranular cracks along grain boundaries and, in severely degraded particles, as intragranular cracks through primary crystallites.[3,15] As shown in Fig. 5b, these cracks create electrically isolated domains that increase internal resistance and expose fresh surfaces to electrolyte attack, accelerating capacity decay.[8] This mechanical legacy poses a distinct challenge for Level II recycling. Cracked secondary particles cannot simply be relithiated and annealed back to their original morphology. Level III strategies that convert polycrystalline secondary particles into single-crystal cathodes address this problem most directly, because single-crystal morphologies eliminate grain boundaries and are therefore intrinsically resistant to intergranular fracture.[23] Temperature accelerates all three of these mechanisms. Studies by Cha *et al.* document that cycling at 60 °C markedly thickens the cathode electrolyte interphase (CEI) layer and intensifies surface reconstruction,[24] while voltage excursions above 4.35 V drive additional rock–salt phase formation in NCA through an HF-mediated dissolution pathway.[25]

These three degradation modes establish a clear hierarchy of material-science challenges for recycling. Restoring lithium stoichiometry alone is necessary but insufficient if cation disorder persists;[26] correcting cation disorder is necessary but insufficient if rock–salt surface reconstruction remains;[27] and even a compositionally and structurally restored particle may underperform if its polycrystalline morphology retains crack-susceptible grain boundaries.[28] This hierarchy directly motivates the organization of recovery strategies as Levels I, II, and III.

2.2 Level I strategies: pyrometallurgy and hydrometallurgy

As summarized in Fig. 3, pyrometallurgy and hydrometallurgy represent the two dominant industrial recycling routes today. Pyrometallurgy processes mixed-chemistry battery feeds at temperatures exceeding 1000 °C, recovering nickel and cobalt as alloys while consigning lithium to the slag phase.[29–31] Its main advantage is robustness and feedstock flexibility. The main limitation, from a structural circularity perspective, is that it does not retain any part of the cathode's crystal structure, and its high thermal output results in a notable carbon footprint. A lightly cycled NMC 811 particle and a heavily degraded one with extensive rock–salt reconstruction are processed the same way, erasing the structural differences that Level II and III strategies aim to exploit.

Hydrometallurgy dissolves spent cathode materials in aqueous acid solutions and selectively precipitates or solvent-extracts the constituent metals as high-purity salts.[32] It operates at far lower energy intensity than pyrometallurgy and recovers lithium efficiently, a critical advantage given tightening supply constraints. However, the dissolution step necessarily destroys the cathode lattice, and the reprecipitation of transition-metal hydroxide precursors for re-calcination into new cathode active material constitutes a full synthesis cycle rather than a recycling cycle. The quality of the re-synthesized material, therefore, depends on the purity and stoichiometry of the recovered precursor, not on the electrochemical quality of the original cathode.[29,33] The use of strong acids also introduces environmental concerns regarding waste management and hazardous by-products. Innovations in green chemistry, including organic acid leachants and environmentally friendly solvents, are reducing the ecological footprint of hydrometallurgical recycling. Still, these represent incremental improvements to a process that remains fundamentally Level I in its recovery logic.

2.3 Level II strategy: direct recycling

Direct recycling departs from the Level I paradigm by seeking to preserve the cathode's layered oxide framework intact throughout the recovery process, restoring electrochemical performance through targeted relithiation and thermal treatment rather than dissolution and re-synthesis.[34] By operating at the phase level rather than the elemental level, direct recycling, in principle, retains all the crystallographic information embedded in the spent cathode, reducing energy consumption relative to full resynthesis.[12] The decisive challenge is that direct recycling must address all three degradation modes described in Section 2 simultaneously. Relithiation corrects lithium deficiency; annealing above the cation-ordering temperature reinstates $R\bar{3}m$ symmetry, but neither step reliably heals rock–salt surface reconstruction or mechanically cracked particle morphologies.[35] The variability of degradation state across a real end-of-life battery pack, reflecting heterogeneous usage histories, further complicates the uniform restoration of electrochemical properties.[35] Direct recycling thus represents a genuine advance in circularity over Level I approaches. Still, its practical effectiveness is bound by the severity of the morphological and surface-chemical damage it cannot fully reverse.

2.4 Level III strategy: upcycling

Upcycling is defined here as any recycling process that not only restores but also deliberately enhances the electrochemical properties of the recovered cathode material relative to its as-

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

manufactured specifications. This distinction matters: a direct recycling process that achieves 98% of original capacity operates at Level II; a process that converts recycled polycrystalline NMC 811 into single-crystal NMC 811 with superior cycling stability operates at Level III, because the recycled product outperforms a conventionally manufactured equivalent.[12] Upcycling strategies include single-crystal conversion from recycled polycrystalline precursors, compositional modification through dopant introduction into the relithiated lattice, and morphological redesign through molten-salt recrystallization. By targeting the grain-boundary fracture and surface instability mechanisms that limit the performance ceiling of polycrystalline Ni-rich cathodes, upcycling addresses the degradation modes that Level II approaches leave unresolved. Significant research is still needed to demonstrate that the performance enhancements achieved in laboratory studies can be maintained at the kilogram scale required for industrial validation.[33]

Substantial progress has been made over the past five years in developing Level II and Level III recycling strategies for Ni-rich cathodes.[36–38] These strategies should be understood not as isolated process options, but as targeted responses to the degradation modes identified above: lithium deficiency and cation disorder, near-surface rock–salt reconstruction, and intergranular fracture. Rather than cataloging all recent technical developments, Section 3 focuses on advances that most directly address these repair targets and critically assesses what each approach has demonstrated and where fundamental

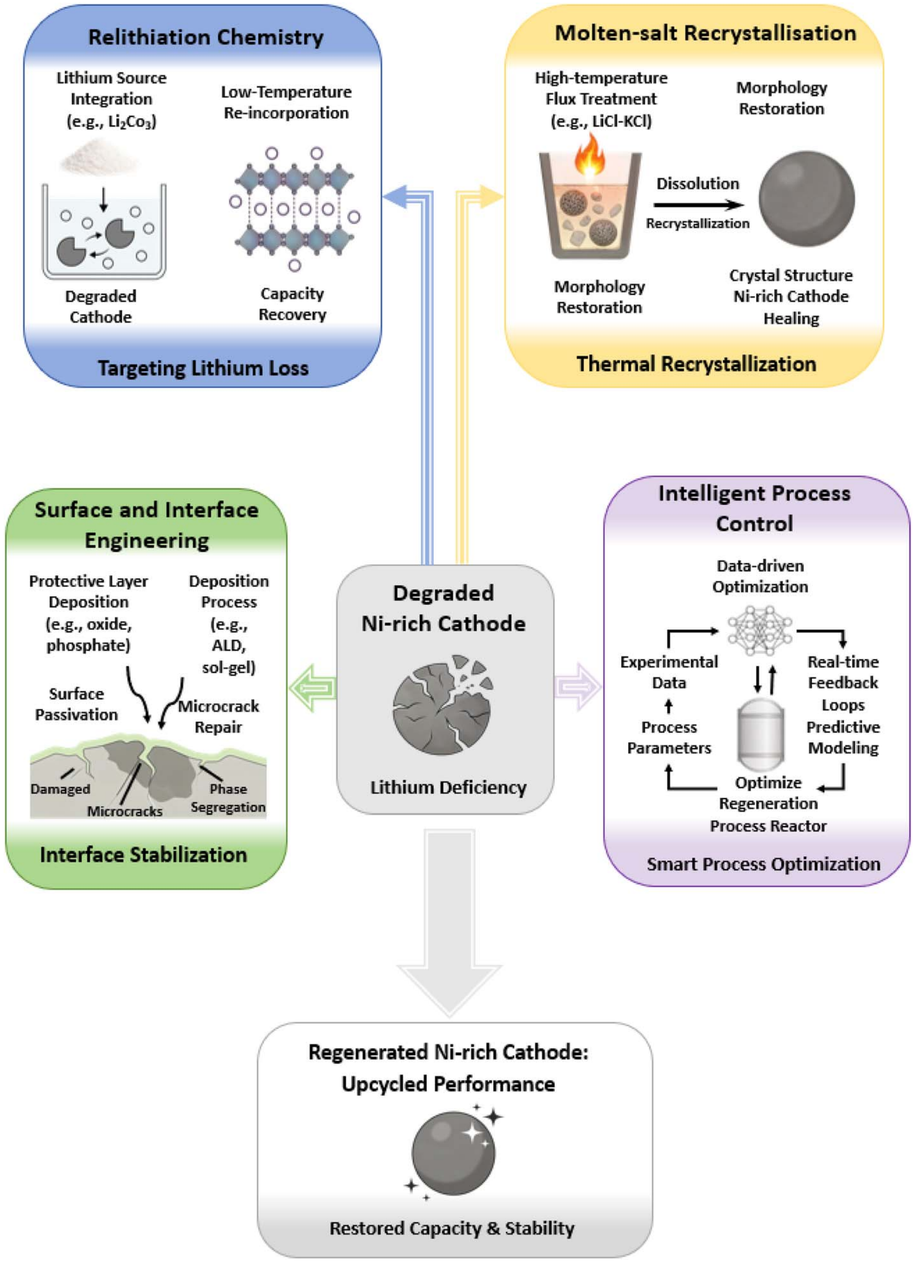

Fig. 6 Emerging regeneration and upcycling strategies for degraded Ni-rich cathodes across multiple length scales.

© 2026 The Author(s). Published by the Royal Society of Chemistry

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

limitations remain. As illustrated in Fig. 6, these advances cluster around four mutually reinforcing themes: relithiation chemistry, molten-salt recrystallization, surface and interface engineering, and intelligent process control.

3 Advances toward structural and functional circularity

3.1 Relithiation chemistry: restoring stoichiometry and lattice order

Correcting lithium deficiency and reversing cation disorder are the two Level II targets most amenable to chemical intervention, and they have driven a diverse body of relithiation research. Conventional thermal lithiation using Li_2CO_3 or LiOH sources above the cation-ordering temperature reinstates $R\bar{3}m$ symmetry and has been demonstrated to recover a substantial fraction of the original capacity.[1] This approach relies on solid-state diffusion at elevated temperatures, however, which limits throughput and can introduce new surface impurities. Representative relithiation windows already span a wide range. Low-temperature aqueous relithiation of spent Ni-rich NMC has been demonstrated at approximately 90 °C,[39] whereas chemically relithiated NMC 622 may still require post-annealing near 720 °C to fully restore the layered structure when deeper crystallographic reconstruction is needed.[40] This wide temperature window highlights why relithiation should be evaluated not only by average lithium recovery, but also by the depth of lattice repair required for a given degradation state.

More mechanistically refined approaches have begun to address these constraints. Redox mediator chemistry, employing compounds such as benzoquinone derivatives, enables solution-phase delivery of both electrons and lithium ions to lithium-deficient cathode lattices, restoring stoichiometry and surface oxidation states without high-temperature treatment.[41] A related topochemical strategy utilizes benzoate-lithium coordination complexes to transport lithium across degraded cathode surfaces *via* a quasi-Grotthuss hopping mechanism, achieving phase uniformity at low temperatures without generating surface impurities.[42] These low-temperature solution-phase approaches are significant because they demonstrate, in principle, that relithiation can be decoupled from high-temperature annealing, a key step toward energy-efficient Level II recycling. Their limitation is that they have so far been demonstrated predominantly on model systems or lightly cycled cathodes; their efficacy on heavily degraded particles with thick rock–salt surface layers and extensive grain-boundary cracking has not yet been systematically established.

Three-in-one solid-phase regeneration, which combines lithium supplementation, dual-anion doping, and lattice repair in a single annealing step, represents a practical approach for simultaneously addressing multiple degradation targets.[23] Artificial Intelligence (AI)-driven matching of degradation signatures to optimized regeneration protocols, based on early-cycle impedance data, provides a data-guided route to tailoring relithiation conditions to the specific degradation state of individual cathode batches.[43] Recent examples show that AI can support recycling at both the diagnostic and process-optimization stages. At the diagnostic stage, federated machine learning has been used to sort 130 retired lithium-ion batteries across five cathode material classes and seven manufacturers, achieving sorting errors of 1% and 3% in homogeneous and heterogeneous recycling settings, respectively.[10] At the process stage, machine-learning-guided multi-objective optimization has been applied to spent-battery black-mass recycling, where a machine learning (ML)-experimental framework identified redox drivers for sulfur/carbon co-roasting and achieved 92.54% selective lithium extraction. These examples suggest that future relithiation and regeneration protocols should move from uniformly applied recipes toward diagnosis-informed, batch-adaptive workflows.[44] The combination of these diagnostic and treatment tools points toward a future in which relithiation is prescribed based on real-time chemical and structural characterization of incoming spent material, rather than applied uniformly across heterogeneous feeds.

3.2 Molten-salt recrystallization: from structural preservation to single-crystal upcycling

Molten-salt processing has emerged as the most powerful demonstrated platform for addressing all three degradation-mode targets, and it is the approach that most clearly bridges Level II and Level III recovery. By dissolving surface rock salt and providing a lithium-rich liquid medium for simultaneous relithiation and lattice recrystallization, low-melting eutectic salt systems can achieve structural restoration at substantially lower temperatures than conventional dry annealing. For example, LiOH–$LiNO_3$ and related nitrate-based eutectic systems have been reported to repair NCM cathodes at approximately 300 °C, whereas other molten-salt systems require higher-temperature steps depending on the salt chemistry and the depth of structural reordering required.[38] Importantly, these conditions also enable the conversion of polycrystalline secondary particles into single-crystal cathodes by dissolving grain boundaries and reprecipitating material as micrometer-scale single crystals,[23,45] which is the only currently demonstrated strategy that directly resolves the intergranular fracture problem. Typical molten-salt processing windows are therefore broad rather than fixed: low-melting LiOH–$LiNO_3$ systems have been used near 300 °C for several hours, LiOH–Li_2CO3 eutectic systems have been reported around 440 °C,[15,46] and some two-step regeneration protocols combine an intermediate molten-salt treatment with high-temperature annealing near 850 °C to complete compositional replenishment and structural reordering.[47]

Single-crystal Ni-rich cathodes (Ni over 90%) produced from recycled polycrystalline precursors *via* this route exhibit capacities exceeding 220 mAh g^{-1} and a markedly reduced susceptibility to microcracking[45] properties that represent a genuine Level III outcome, where the recycled product outperforms a conventionally manufactured polycrystalline equivalent. Additional process innovations within the molten-salt platform further extend its capabilities. Boric acid-etched precursors generate *in situ* $LiBO_2$ surface layers that passivate the cathode

© 2026 The Author(s). Published by the Royal Society of Chemistry

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM. This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

against electrolyte decomposition at elevated voltage.[11] Hydrogen reduction before molten-salt processing selectively reduces Ni^{2+}/Co^{3+} to divalent states, preventing the formation of rock–salt and spinel byproducts during recrystallisation.[48] Mild eutectic mixtures, such as $LiNO_3$–$NaNO_3$, can achieve surface restoration at ambient pressure without inert atmospheres, thereby lowering the technical barriers to scale-up.[49]

Molten-salt upcycling still faces significant challenges on the path to industrial deployment, however. Process yield and single-crystal size distribution are sensitive to salt composition, temperature profile, and the morphological heterogeneity of the input material, all parameters that are difficult to control when processing real end-of-life battery black mass with variable degradation states. The cost of lithium salts at the quantities required for industrial-scale molten-salt processing also requires careful techno-economic justification, particularly if the recovered lithium from the spent cathode does not fully offset the process lithium demand. These constraints indicate that molten-salt recrystallization should be treated as a high-value but condition-dependent Level III route rather than a universally applicable solution. Its practical viability will depend on whether salt recovery, lithium-source utilization, corrosion management, product-size control, and battery-grade qualification can be maintained at scale. This limitation is particularly important for real black-mass feedstocks, where impurity levels, cathode chemistries, and degradation histories vary substantially across cells and packs.[38,46,50]

3.3 Surface and interface engineering: protecting the restored architecture

Bulk relithiation and recrystallization can restore average stoichiometry and long-range structural order, but, by themselves, they do not guarantee durable electrochemical behavior. A structurally restored Level II or Level III cathode may still fail prematurely if its surface remains vulnerable to electrolyte attack, oxygen release, transition-metal dissolution, or impedance growth. Surface and interface engineering, therefore, serves as a necessary complement to relithiation and recrystallization rather than a standalone recycling approach.

Atomic Layer Deposition (ALD) of nanoscale coatings, such as Al_2O_3 and ZrO_2, yields conformal films under 5 nm thick that reduce interfacial resistance while maintaining electrolyte access to the active material.[51] Amorphous $LiBO_2$ coatings serve as chemically inert barriers against HF attack and oxygen evolution during aggressive cycling.[11] Core-shell architectures with Ni-rich cores and $LiMn_2O_4$ or $LiCoPO_4$ shells promote structural reversibility and mitigate thermal instability under high-voltage operation.[52] At the electrode level, polyimide-siloxane binders that capture dissolved transition metals prevent cathode fading and structural delamination,[53] while lattice doping with W^{6+} strengthens MO_6 octahedra and resists collapse during deep charging.[54]

The evaluative question for these surface strategies is whether the protection they provide is durable enough to justify the additional processing steps and cost relative to simply producing a new cathode particle. The answer depends critically on the baseline performance of the recycled core: a surface coating applied to a thoroughly relithiated, single-crystal core operating at Level III has a very different value proposition from the same coating applied to a barely treated Level I hydrometallurgical output.

3.4 Selective metal recovery: enabling high-purity inputs for structural recycling

The effectiveness of Level II and Level III strategies depends on the purity and compositional precision of the feeds they process. Selective transition-metal recovery therefore functions as a critical upstream enabler for structural recycling, not merely a standalone Level I objective. Ligand-assisted leaching with chelating agents such as pyrophosphates or phosphonic acids achieves over 95% recovery of Ni, Co, and Mn with minimal cross-contamination, providing precursor streams suitable for direct reprecipitation into compositionally accurate NMC or NCA precursors.[55] Sulfated-reduction roasting followed by stage-wise selective leaching enables sequential high-purity metal separation.[55] Solvent-assisted stripping with tailored extractants, such as di-(2-ethylhexyl) phosphinic acid (P227), achieves over 90% recovery of critical metals while preventing lithium loss.[56] Deep eutectic solvent-based co-precipitation streamlines precursor synthesis, eliminating filtration steps and enabling over 90% purity in recovered Ni–Co–Mn mixtures.[48]

These advanced separation methods are currently used primarily as higher-selectivity alternatives to conventional hydrometallurgy, that is, still as Level I tools. Their potential to serve as precision feed preparation for Level II or III processing represents an underexplored integration opportunity. Combining selective leaching and reprecipitation with direct relithiation or molten-salt upcycling could allow a two-stage process that recovers the structural value of lightly degraded particles and the elemental value of heavily degraded ones from the same battery feed.

4 Feasibility of scaling up structural and functional recycling

4.1 Techno-economic considerations

The scalability of Level II and III recycling hinges on bridging the gap between demonstrated laboratory performance and industrially viable economics. As summarized in Fig. 7, feasibility is governed by coordinated progress across technology readiness, process economics, regulatory compliance, and market integration. With EV adoption driving LIB recycling demand forecast to exceed 5 million metric tons by 2030,[13] the economic case for structural recycling is strengthening. Direct recycling methods employing molten salts and solid-state relithiation are emerging as candidates for scalable solutions: reciprocal ternary molten salt (RTMS) systems can regenerate cathodes such as NMC 622 with performance metrics rivalling those of pristine materials.[50,57,58] In contrast, low-temperature solvated-electron processes regenerate cathode material under

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

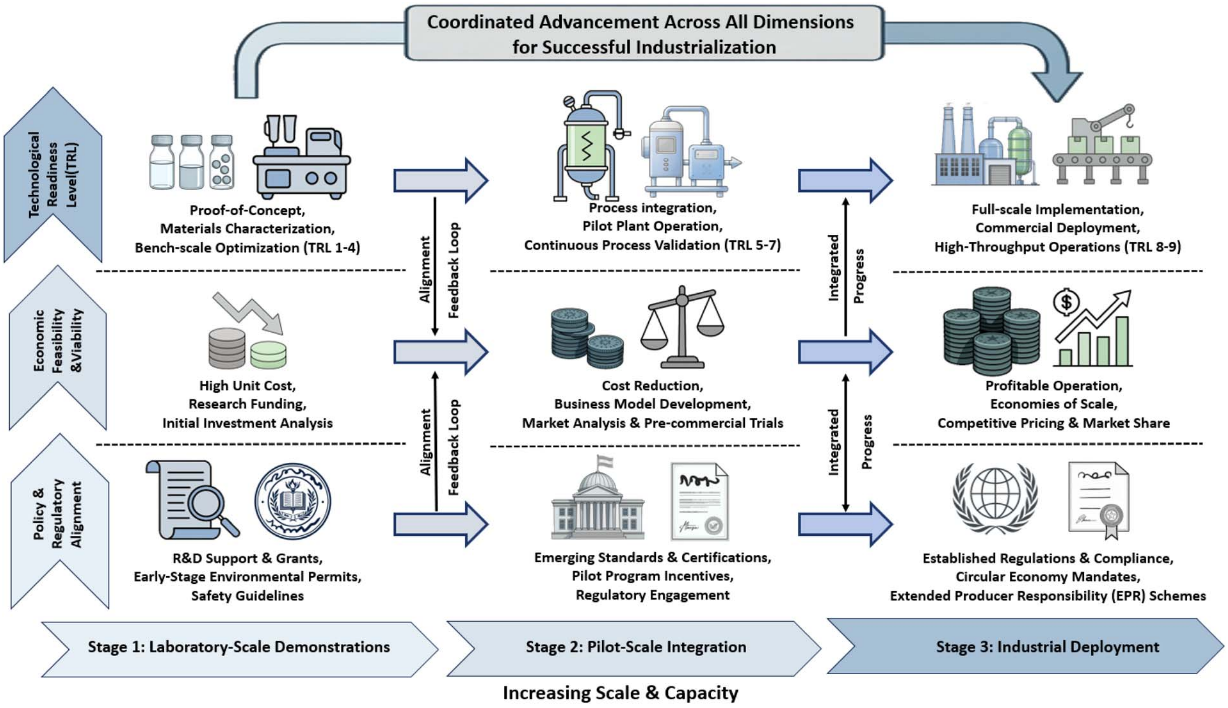

Fig. 7 Multidimensional pathway for scaling up Ni-rich cathode recycling from laboratory demonstration to industrial deployment.

ambient conditions within minutes, dramatically reducing energy demand.[59]

Techno-economic assessments indicate that Level II recycling economics are most compelling when material value retention is maximized, and reagent consumption is minimized. Hydrometallurgical co-extraction of Mn–Co–Ni and lithium, achieving over 98% recovery while enabling reuse without full separation, reduces both reagent use and operational steps relative to conventional approaches.[60,61] Integrating electrodialysis for lithium extraction has reduced reagent costs and emissions,[62] and vacuum membrane distillation has demonstrated up to 32% lower energy usage for lithium recovery.[63] Transforming polycrystalline cathodes into single-crystal NMCs improves cycling stability, achieving 94% retention after 500 cycles, while also improving per-kilogram cost metrics.[64] Laser-assisted Ni-doping regeneration and nanobubble-enhanced leaching have also delivered cost and performance improvements,[65,66] with economic analyses suggesting potential gains of up to $20,000 USD per ton from optimized upcycling integrated into upstream battery production workflows.[14] At the route level, recent cost-modeling reviews report indicative profit margins of approximately US $0.4–3.3 kg^{-1} for hydrometallurgy, US $0.5–4.0 kg^{-1} for pyrometallurgy, and US $2.0–14.4 kg^{-1} for direct recycling, although these values vary strongly with chemistry, scale, process assumptions, cost categories, and recovered product type.[67] These data support the economic promise of Level II and III routes, but also reinforce the need for transparent and scale-sensitive cost models when comparing them with established Level I pathways.

4.2 Policy and regulatory context

While a detailed policy analysis lies outside the scope of a chemistry perspective, the regulatory environment is shaping which recycling technologies receive development investment and therefore warrants brief consideration. The EU Battery Regulation's minimum recycled-content mandates for cobalt, lithium, and nickel from 2030 directly incentivize closed-loop structural recycling over open-loop elemental recovery, because meeting these thresholds with battery-grade material requires the compositional precision and structural quality that only Level II and III processes can reliably deliver.[68] China's extended producer responsibility framework has already increased LIB collection rates and raised the market value of recycled metals,[69] creating commercial pull for advanced recycling at scale. The emerging "3L" policy standard (less energy, less emissions, less cost) in both the EU and Asia aligns regulatory incentives with the low-temperature,[70] structure-preserving processes this perspective identifies as scientifically most promising. What remains absent from current policy frameworks is a mechanism for rewarding the electrochemical quality, rather than merely the mass, of recovered cathode material; establishing such a quality-based standard would be a powerful instrument for accelerating the transition from Level I to Level II and III recycling. At the same time, policy incentives should not imply that Level III recycling is always the preferred route.[71] Higher-value recovery generally requires additional diagnostic, relithiation, recrystallization, compositional tuning, and quality-control steps, which increase process complexity and cost. Recent cost-modeling studies suggest that direct recycling can be economically attractive under favorable

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

assumptions, but reported outcomes remain highly sensitive to system boundaries, product assumptions, and whether transportation, pack disassembly, labor, and capital expenditure are included.[39,40] Therefore, the transition from Level I to Level II or III should be evaluated not only by the nominal value of the recovered cathode, but also by scale-aware techno-economic performance and battery-grade qualification.

5 Unsolved problems and future directions

5.1 The heterogeneity problem: recycling real end-of-life material

The advances described in Section 3 demonstrate genuine progress toward structural and functional circularity for Ni-rich cathodes, but they also expose the inherent limitations of the three-level framework. The framework is intended as a value-retention map rather than a universal process hierarchy: Level II and Level III routes are scientifically attractive only when the additional complexity of diagnosis, defect repair, recrystallization, and qualification is justified by a measurable gain in cathode value.[46,57,67] Therefore, the unresolved problems in this field can be organized into three high-impact priority directions. The first is adaptive sorting and diagnosis for heterogeneous end-of-life feedstocks. The second is spatially uniform defect repair, including quantitative control of lithium redistribution, cation ordering, and the removal of surface reconstruction. The third is the integration of structure-preserving regeneration with scale-aware process control, quality assurance, and battery-grade qualification.

The first priority, adaptive sorting and diagnosis, arises from the gap between current laboratory demonstrations and the heterogeneity of real end-of-life cathode material. Laboratory studies of direct recycling and upcycling are almost invariably performed on homogeneous model samples, cathodes cycled under controlled conditions to a defined state of health, whereas a real battery pack contains particles spanning a wide distribution of degradation states, degradation modes, and even cathode chemistries.[72] Adaptive recycling protocols that can sort and treat incoming material based on real-time characterization of its degradation state are, therefore, a critical unsolved problem. Internet of Things (IoT)-enabled monitoring systems that track individual cell histories and pre-sort end-of-life material by degradation profile offer a promising direction.[73] Federated machine learning provides a more concrete example of how such a diagnosis could be implemented across distributed stakeholders: one recent study classified 130 retired lithium-ion batteries spanning five cathode material classes and seven manufacturers without requiring collaborators to share proprietary operating data, achieving cathode-sorting errors of 1% and 3% under homogeneous and heterogeneous recycling settings, respectively.[10] This result is important because Level II and Level III recycling are cathode-specific; inaccurate sorting can send chemically incompatible or excessively degraded material into a treatment route for which it is not suited. Nevertheless, no study has yet demonstrated that a single Level II or III process can reliably restore electrochemical performance across the full distribution of degradation states present in a commercial EV pack, and this remains arguably the most important unresolved question in the field.

5.2 The relithiation and defect-uniformity problem

The second priority is spatially uniform defect repair. Existing relithiation techniques, whether thermal, solution-phase, or molten-salt-mediated, can restore average lithium stoichiometry but do not necessarily guarantee uniform lithium redistribution within and across cathode particles.[37,46] Lithium-deficient regions that are not fully reactivated by the relithiation reagent exhibit local capacity heterogeneity, which accelerates subsequent degradation. This problem is compounded by the spatially variable thickness of the rock–salt surface layer across a real particle surface,[3] whose dissolution is a prerequisite for effective relithiation. Future research should prioritize combining hydrothermal relithiation with defect management to enhance lithium distribution and bolster structural stability,[46] alongside phase-selective recovery approaches that remove inactive phases while preserving the active layered domains.[74] Piezoelectric coatings that mitigate stress accumulation during relithiation offer a complementary route to stabilizing cathode surfaces throughout the restoration process.[75]

5.3 Scale-aware Level III upcycling and battery-grade qualification

The third priority is to translate Level III upcycling from proof-of-concept material redesign into scale-aware, battery-grade cathode manufacturing. The most ambitious frontier is the use of recycled Ni-rich cathode material not merely as a feedstock for reproducing the original composition, but as a basis for synthesizing next-generation cathode architectures. Recent studies have shown that NCM811 synthesized from spent NCM111 can outperform commercial counterparts in energy capacity and cycling stability.[76] Other studies have further shown that recycled battery-derived metal streams can be redirected toward alternative high-voltage cathode chemistries, such as $LiFe_{0.5}Mn_{0.5}PO_4$, when additional compositional sources are introduced.[77] These results establish a proof of concept but also highlight a fundamental tension: designing toward a target composition requires precise control of input elemental ratios, which, in turn, requires the kind of selective separation currently most reliably achieved by Level I hydrometallurgical processing. Resolving this tension, that is, developing hybrid processes that preserve structural value where possible while enabling compositional redesign where desired, is one of the most important integration challenges facing the field. For industrial implementation, this integration challenge must be evaluated against qualification-relevant metrics rather than solely laboratory half-cell performance. A Level III product must meet reproducible specifications in composition, residual impurity content, particle-size distribution, tap density, moisture sensitivity, electrode-processing compatibility, full-cell performance, and safety. Future studies should therefore

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

report not only the electrochemical performance of the upcycled cathode, but also process yield, reagent recovery, batch-to-batch reproducibility, impurity tolerance, and qualification-relevant full-cell data.

5.4 Life-cycle and sustainability implications

Life-cycle assessment (LCA) studies generally suggest that Level II and Level III recycling can reduce energy use, carbon emissions, and secondary waste relative to full elemental recovery and resynthesis, particularly when the recovered cathode structure or function can be directly reused. Direct cathode regeneration avoids the intensive thermal or chemical treatments required for full resynthesis, reducing greenhouse gas emissions and secondary waste.[78] A comprehensive LCA of LIBs found that most environmental impacts arise from production and end-of-life (EoL) treatment, both of which are significantly mitigated when upcycling is applied.[72,79] Electrochemical-impedance-based state-of-health diagnostics can further support this decision-making process by identifying cells whose degradation profiles are suitable for higher-value regeneration or upcycling, thereby avoiding unnecessary high-intensity processing of severely damaged materials.[80] Upcycling degraded cathode material instead of refining raw minerals also reduces the embedded energy and emissions associated with mining and high-temperature synthesis.[81] Data-driven models now enable prediction of battery cycle life from as few as 10 early cycles with under 12% error,[82] creating opportunities to route batteries with the most favorable structural profiles toward Level III upcycling while directing those with extensive morphological damage toward Level I or second-life applications.

5.5 The collaboration imperative

None of the scientific advances described above will translate into industrial impact without structural changes in how battery value chains are organized. Achieving true structural circularity requires that recyclers receive end-of-life batteries with reliable provenance data, that manufacturers design cells for disassembly, and that cathode producers accept recycled active material as a direct input rather than a commodity precursor. Human–robot collaborative disassembly systems being piloted for EV battery recycling represent one enabling technology,[83] but the deeper need is for standardized battery designs and passports that encode the electrochemical history of individual cells.[84] Partnerships across the value chain, from mining through cathode production to recycling, are essential for securing sustainable material flows in a rapidly growing EV market,[85] and academic-industry collaboration is accelerating the translation of advanced AI, machine learning, and green chemistry insights into practical applications.[86] Coordinated policy, standardization, and corporate commitment to circular design are all necessary to make structural and functional cathode recycling mainstream and scalable.[87] In this context, AI is most useful when heterogeneous feedstocks make fixed treatment recipes unreliable and when recycling performance depends on rapid mapping from degradation signatures to repair protocols.

6 Conclusions

This Perspective argues that Ni-rich cathode recycling cannot be fully addressed through elemental recovery alone. Surface rock–salt reconstruction, cation disorder, and intergranular fracture progressively destroy the crystal architecture that gives these materials their electrochemical value. Level I processes can recover metal inventory, but they do not recover this architectural information. The transition to Level II structural preservation and Level III functional restoration is therefore not an incremental improvement. It is a necessary reorientation of recycling research and industrial practice.

The most important advances toward this reorientation include molten-salt recrystallization, redox-mediated relithiation, surface and interface engineering, and AI-guided process control. Molten-salt routes can couple surface repair, lithium replenishment, and recrystallization, and in some cases enable single-crystal conversion. Redox-mediated and solution-phase relithiation methods offer lower-temperature routes to stoichiometric repair. Surface and interface engineering protects the restored architecture from renewed electrolyte attack. AI-guided diagnosis and process optimization provide a route toward batch-adaptive treatment of heterogeneous end-of-life feedstocks. Together, these approaches show that cathode recycling is evolving from a primarily metallurgical operation into a materials-science and chemical-engineering discipline.

Several barriers still limit this transition. Real end-of-life cathode streams are chemically and structurally heterogeneous. Lithium redistribution during relithiation remains difficult to control uniformly across particles and batches. Level III upcycling also requires precise compositional control, battery-grade qualification, and techno-economic justification at scale. Addressing these challenges will require new chemistry, but it will also require better institutional infrastructure, including standardized cell designs, digital material passports, and quality-based regulatory standards that reward electrochemical performance rather than mass recovery alone.

The broader opportunity is to treat the crystal architecture of a spent cathode as a resource to be repaired or upgraded, not as an ore to be dissolved. Recycling success should therefore be measured not only in tonnes of metal recovered, but also in the electrochemical quality of the cathode materials returned to productive use.

Author contributions

Y. Huang and G. Li contributed to the conception and design of the review. Y. Huang drafted the manuscript. H. Xu, X. Han, Y. Zeng, and G. Li revised the manuscript.

Conflicts of interest

There are no conflicts to declare.

© 2026 The Author(s). Published by the Royal Society of Chemistry

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

Data availability

There is no additional data associated with this article.

Acknowledgements

This work was financially supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) through the Discovery Grant Program (RGPIN-2020-05184) and the NSERC Alliance (ALLRP 586704-2023). The authors acknowledge support from CanmetMATERIALS through the Natural Resources Canada Critical Minerals Program.

References

1 W. Mrozik, M. A. Rajaeifar, O. Heidrich and P. Christensen, Environmental Impacts, Pollution Sources and Pathways of Spent Lithium-Ion Batteries, *Energy Environ. Sci.*, 2021, **14**(12), 6099–6121, DOI: 10.1039/D1EE00691F.

2 Y.-S. Chen, R. Dominko, M. Marczewski and W. Wieczorek, Optimizing High-Energy Lithium-Ion Batteries: A Review of Single Crystalline and Polycrystalline Nickel-Rich Layered Cathode Materials: Performance, Synthesis and Modification, *Appl. Phys. A*, 2024, **130**(10), 740, DOI: 10.1007/s00339-024-07897-7.

3 T. Li, X.-Z. Yuan, L. Zhang, D. Song, K. Shi and C. Bock, Degradation Mechanisms and Mitigation Strategies of Nickel-Rich NMC-Based Lithium-Ion Batteries, *Electrochem. Energy Rev.*, 2020, **3**(1), 43–80, DOI: 10.1007/s41918-019-00053-3.

4 J. Song, L. Huang, G. Yang, T. Liu, S. Liu, G. Cong, Y. Huang, Z. Liu, X. Gao and L. Geng, Regulating Grind-Induced Lattice Distortion for Nickel-Rich Cathodes by Annealing, *Small Methods*, 2024, **8**(3), 2301400, DOI: 10.1002/smtd.202301400.

5 X. Chang, M. Fan, C.-F. Gu, W. He, Q. Meng, L. Wan and Y.-G. Guo, Selective Extraction of Transition Metals from Spent LiNixCoyMn1-x-yO2 Cathode *via* Regulation of Coordination Environment, *Angew. Chem.*, 2022, 2202558, DOI: 10.1002/anie.202202558.

6 S. K. Hussain and J. H. Bang, Recent Progress in Co-Free, Ni-Rich Cathode Materials for Lithium-Ion Batteries, *Bull. Korean Chem. Soc.*, 2024, **45**(1), 4–15, DOI: 10.1002/bkcs.12799.

7 H. Choi, A. R. Schuer, H. Moon, G. Melinte, G.-T. Kim, J. Asenbauer, A. Kazzazi, M. Kuenzel and S. Passerini, Is Cobalt in Li-Rich Layered Oxides for Li-Ion Batteries Necessary?, *ChemElectroChem*, 2024, **11**, e202400391, DOI: 10.1002/celc.202400391.

8 E. Elmaataouy, A. Chari, A. E. Bendali, M. Tayoury, R. Amine, M. Aqil, G. Xu, T. Liu, J. Alami and M. Dahbi, LiNi0.8Fe0.1Al0.1O2 as a Cobalt-Free Cathode Material with High Capacity and High Capability for Lithium-Ion Batteries, *Batteries*, 2022, **9**(1), 23, DOI: 10.3390/batteries9010023.

9 C. Earl, I. H. Shah, S. Cook and C. R. Cheeseman, Environmental Sustainability and Supply Resilience of Cobalt, *Sustainability*, 2022, **14**(7), 4124, DOI: 10.3390/su14074124.

10 S. Tao, H. Liu, C. Sun, H. Ji, G. Ji, Z. Han, R. Gao, J. Ma, R. Ma, Y. Chen, S. Fu, Y. Wang, Y. Sun, Y. Rong, X. Zhang, G. Zhou and H. Sun, Collaborative and Privacy-Preserving Retired Battery Sorting for Profitable Direct Recycling *via* Federated Machine Learning, *Nat. Commun.*, 2023, **14**(1), 8032, DOI: 10.1038/s41467-023-43883-y.

11 Z. Guo, X. Shi, L. Cao, J. Zhang, X. Zhang, J. Yao, Y.-J. Cheng and Y. Xia, In Situ Formation of a LiBO2 Coating Layer and Spinel Phase for Ni-Rich Cathode Materials from a Boric Acid-Etched Precursor, *ACS Appl. Mater. Interfaces*, 2024, **16**(1), 731–741, DOI: 10.1021/acsami.3c14342.

12 Y. Zhang, N. Yao, X. Tang, H. Wang, M. Zhang, Z. Wang, A. Shao, J. Liu, L. Cheng, Y. Guo and Y. Ma, Upcycling of High-Rate Ni-Rich Cathodes through Intrinsic Structural Features, *Adv. Energy Mater.*, 2024, 2402918, DOI: 10.1002/aenm.202402918.

13 M. M. Gnutzmann, A. Makvandi, J. Buchmann, M. Winter and J. Kasnatscheew, Understanding Solid-State Direct Recycling of Ni-Rich Layered Positive Electrodes, *Meet. Abstr.*, 2024, (5), 554, DOI: 10.1149/MA2024-025554mtgabs.

14 Y. Sun, H. Yang, J. Li, J. Li, X. Zhuge, Y. Ren and Z. Ding, A Large Volume and Low Energy Consumption Recycling Strategy for LiNi0.6Co0.2Mn0.2O2 from Spent Ternary Lithium-Ion Batteries, *J. Power Sources*, 2024, **602**, 234407, DOI: 10.1016/j.jpowsour.2024.234407.

15 P. Teichert, G. G. Eshetu, H. Jahnke and E. Figgemeier, Degradation and Aging Routes of Ni-Rich Cathode Based Li-Ion Batteries, *Batteries*, 2020, **6**(1), 8, DOI: 10.3390/batteries6010008.

16 H.-H. Ryu, H.-W. Lim, S. G. Lee and Y.-K. Sun, Near-Surface Reconstruction in Ni-Rich Layered Cathodes for High-Performance Lithium-Ion Batteries, *Nat. Energy*, 2024, **9**(1), 47–56, DOI: 10.1038/s41560-023-01403-8.

17 W. Wang, Y. Shi, P. Li, R. Wang, F. Ye, X. Zhang, W. Li, Z. Wang, C. Xu, D. Xu, Q. Xu and X. Cui, Rational Rock–Salt Phase Engineering of a Nickel-Rich Layered Cathode Interface for Enhanced Rate and Cycling Stability, *Energy Environ. Sci.*, 2024, **17**(12), 4283–4294, DOI: 10.1039/D3EE04110G.

18 L. Zou, W. Zhao, Z. Liu, H. Jia, J. Zheng, G. Wang, Y. Yang, J.-G. Zhang and C. Wang, Revealing Cycling Rate-Dependent Structure Evolution in Ni-Rich Layered Cathode Materials, *ACS Energy Lett.*, 2018, **3**(10), 2433–2440, DOI: 10.1021/acsenergylett.8b01490.

19 J. Shen, Regulating the Li/Ni Mixing Ratio to Enhance Transition Metal-Lattice Oxygen Interaction for Achieving Long Life Lithium-Rich Layered Oxides, *Angew. Chem., Int. Ed.*, 2026, e19458- Wiley Online Library. https://onlinelibrary-wiley-com.login.ezproxy.library.ualberta.ca/doi/10.1002/anie.202519458.

20 Z. Huang, Z. Chen, M. Yang, M. Chu, Z. Li, S. Deng, L. He, L. Jin, R. E. Dunin-Borkowski, R. Wang, J. Wang, T. Yang and Y. Xiao, Insights into the Defect-Driven Heterogeneous Structural Evolution of Ni-Rich Layered Cathodes in

© 2026 The Author(s). Published by the Royal Society of Chemistry

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

Lithium-Ion Batteries, *Energy Environ. Sci.*, 2024, **17**(16), 5876–5891, DOI: 10.1039/D4EE01777C.

21 Y. Zhu, Y. Huang, R. Du, M. Tang, B. Wang and J. Zhang, Effect of Ni2+ on Lithium-Ion Diffusion in Layered LiNi1−x−yMnxCoyO2 Materials, *Crystals*, 2021, **11**(5), 50465, DOI: 10.3390/cryst11050465.

22 H. Ronduda, M. Zybert, A. Szczęsna-Chrzan, T. Trzeciak, A. Ostrowski, D. Szymański, W. Wieczorek, W. Raróg-Pilecka and M. Marcinek, On the Sensitivity of the Ni-Rich Layered Cathode Materials for Li-Ion Batteries to the Different Calcination Conditions, *Nanomaterials*, 2020, **10**(10), 2018, DOI: 10.3390/nano10102018.

23 S. Fang, S. Zhang, L. Ni, G. Zou, H. Hou, H. Liu, W. Deng and X. Ji, Electrochemically Engineering a Single-Crystal Nickel-Rich Layered Cathode, *Inorg. Chem.*, 2023, **62**(11), 4514–4524, DOI: 10.1021/acs.inorgchem.2c04284.

24 H. Cha, J. Hwang, T. Lee, J. Kang, M. Park and J. Cho, Exploring the Degradation Pathways of a Nickel-Rich Cathode during High-Temperature Storage in High-Energy Lithium-Ion Batteries, *J. Mater. Chem. A*, 2023, **11**(28), 15475–15481, DOI: 10.1039/D3TA01991H.

25 Z. Wang, M. M. Rao, J. Li, C. Ye, Z. Liu, Q. Xu, X. Jin, R. Du, Q. Xie, W. Luo, W. Li, Y. Qiu, Z. Wang, M. M. Rao, J. Li, C. Ye, Z. Liu, Q. Xu, X. Jin, R. Du, Q. Xie, W. Luo, W. Li and Y. Qiu, Understanding the Mechanism of Cycling Degradation and Novel Strategy to Stabilize the Cycling Performance of Graphite/LiCoO2 Battery at High Voltage, *J. Electroanal. Chem.*, 2019, **851**, 113411, DOI: 10.1016/j.jelechem.2019.113411.

26 M. Ahangari, B. Szalai, J. Lujan, M. Zhou and H. Luo, Advancements and Challenges in High-Capacity Ni-Rich Cathode Materials for Lithium-Ion Batteries, *Materials*, 2024, **17**(4), 040801, DOI: 10.3390/ma17040801.

27 W. Zhao, M. Si, K. Wang, E. Brack, Z. Zhang, X. Fan and C. Battaglia, Electrolyte Optimization to Improve the High-Voltage Operation of Single-Crystal LiNi0.83Co0.11Mn0.06O2 in Lithium-Ion Batteries, *Batteries*, 2023, **9**(11), 528, DOI: 10.3390/batteries9110528.

28 G. Qu, X. Chen, F. Yang, Z. Yang, G. Liu, J. Lee, R. Fong, J. Tian, R. Wen, C. Wang and Y. Huang, Boosting Ni-Rich Cathode Stability Through Grain Boundary Strengthening and Phase Transition Degradation Suppression, *Small*, 2025, **21**(23), 2502609, DOI: 10.1002/smll.202502609.

29 K. Davis and G. P. Demopoulos, Effective Upcycling of NMC 111 to NMC 622 Cathodes by Hydrothermal Relithiation and Ni-Enriching Annealing, *Next Energy*, 2024, **4**, 100122, DOI: 10.1016/j.nxener.2024.100122.

30 V. Marcinov, D. Oráč, J. Klimko, Z. Takáčová, J. Piroškova and O. Jankovský, Selective Precipitation of REE-Rich Aluminum Phosphate with Low Lithium Losses from Lithium Enriched Slag Leachate, *Materials*, 2024, **17**(20), 5113, DOI: 10.3390/ma17205113.

31 S. Arnold, J. G. A. Ruthes, C. Kim and V. Presser, Electrochemical Recycling of Lithium-Ion Batteries: Advancements and Future Directions, *EcoMat*, 2024, e12494, DOI: 10.1002/eom2.12494.

32 S. A. Jose, J. L. Stoll, T. Smith, C. Jackson, T. Dieleman, E. Leath, N. Eastwood and P. L. Menezes, Critical Review of Lithium Recovery Methods: Advancements, Challenges, and Future Directions, *Processes*, 2024, **12**(10), 2203, DOI: 10.3390/pr12102203.

33 M. C. Etude, A. I. Ikeuba, C. N. Njoku, E. Yakubu, H. C. Uzoma, C. E. Mgbemere and D. I. Udunwa, Recycling Lithium-Ion Batteries: A Review of Current Status and Future Directions, *Sustain. Chem. One World*, 2024, **4**, 100027, DOI: 10.1016/j.scowo.2024.100027.

34 S. Bhattacharyya, S. Roy and R. Vajtai, Emerging Processes for Sustainable Li-Ion Battery Cathode Recycling, *Small*, 2025, **21**(28), 2400557, DOI: 10.1002/smll.202400557.

35 U. Kar, S. Nili, E. Mends, E. Vahidi and P. Chu, A Review and Environmental Impact Analysis on the Current State of Froth Flotation on Recycling of E-Wastes, *Resour., Conserv. Recycl.*, 2025, **212**, 107967, DOI: 10.1016/j.resconrec.2024.107967.

36 Y. Lan, X. Li, G. Zhou, W. Yao, H.-M. Cheng and Y. Tang, Direct Regenerating Cathode Materials from Spent Lithium-Ion Batteries, *Adv. Sci.*, 2024, **11**(1), 2304425, DOI: 10.1002/advs.202304425.

37 Y. Ji, E. E. Kpodzro, C. T. Jafvert, F. Zhao, Y. Ji, E. E. Kpodzro, C. T. Jafvert and F. Zhao, Direct Recycling Technologies of Cathode in Spent Lithium-Ion Batteries, *Clean Technol. Recycl.*, 2021, **1**(2), 124–151, DOI: 10.3934/ctr.2021007.

38 Z. Qin, Y. Zhang, W. Luo, T. Zhang, T. Wang, L. Ni, H. Wang, N. Zhang, X. Liu, J. Zhou and G. Chen, A Universal Molten Salt Method for Direct Upcycling of Spent Ni-Rich Cathode towards Single-Crystalline Li-Rich Cathode, *Angew. Chem.*, 2023, **135**(25), e202218672, DOI: 10.1002/ange.202218672.

39 N. N. Tham, J. Zhang, H. R. Tan, W. Wang, A. Sng, Y. Y. K. Hnin, F. Wei, Y. Ren and Z. Liu, Low-Temperature Direct Regeneration of Spent Nickel-Rich NMC *via* Aqueous Relithiation, *ACS Appl. Mater. Interfaces*, 2026, **18**(2), 3696–3704, DOI: 10.1021/acsami.5c15726.

40 J. I. Preimesberger, C. K. Kirwa, E. Allen, N. S. Dutta, E. Wang, F. Dogan, P. Walker, Y. Bai, K. Z. Pupek, L. Stanley, T. L. Kinnibrugh, M. Nisbet, T. T. Fister, H. Luo and J. E. Coyle, Effect of Annealing on Direct Recycled NMC Cathodes, *Chem. Mater.*, **37**(23), 9406–9419, DOI: 10.1021/acs.chemmater.5c01824.

41 C. K. Kirwa, J. Coyle and H. Luo, Direct Recycling of End-of-Life Lithium-Ion Battery Cathode Materials through Redox Mediator Chemistry, *Meet. Abstr.*, 2023, **1**(4), 853, DOI: 10.1149/MA2023-014853mtgabs.

42 Y. He, K. Jia, Z. Piao, Z. Cao, M. Zhang, P. Li, Z. Li, Z. Jiang, G. Yang, H. Xi, G. Zhou, W. Tang, Z. Qu, R. V. Kumar, S. Ding and K. Xi, Li+ Quasi-Grotthuss Topochemistry Transport Enables Direct Regeneration of Spent Lithium-Ion Battery Cathodes, *Angew. Chem., Int. Ed.*, 2025, **64**(13), e202422610, DOI: 10.1002/anie.202422610.

43 M. Alyoubi, I. Ali and A. M. Abdelkader, Optimising the Regeneration Process of Spent Lithium-cobalt Oxide Cathode through Performance Analysis Model, *J. Energy Storage*, 2025, **110**, 115132, DOI: 10.1016/j.est.2024.115132.

44 J. Qu, B. Zhang, R. Jiang, J. Huang, M. Li, S. Zheng, W. Yu, P. Sun, Y. Xu, Y. Hou, H. Lin and Z. Yu, Machine Learning-

© 2026 The Author(s). Published by the Royal Society of Chemistry

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

Guided Multi-Objective Optimization for Selective Lithium Extraction and Valuable Metal Reuse from Spent Lithium-Ion Batteries, *Chem. Eng. J.*, 2025, **525**, 169978, DOI: 10.1016/j.cej.2025.169978.

45 S. Sun, E. Fan, H. Wang, X. Lv, X. Zhang, R. Chen, F. Wu and L. Li, In Situ Constructed Spinel Layer Stabilized Upcycled LiCoO2 for High Performance Lithium-Ion Batteries, *Small*, 2024, **20**(34), 2401089, DOI: 10.1002/smll.202401089.

46 M. Huang, M. Wang, L. Yang, Z. Wang, H. Yu, K. Chen, F. Han, L. Chen, C. Xu, L. Wang, P. Shao and X. Luo, Direct Regeneration of Spent Lithium-Ion Battery Cathodes: From Theoretical Study to Production Practice, *Nano-Micro Lett.*, 2024, **16**(1), 207, DOI: 10.1007/s40820-024-01434-0.

47 G. Mehboob, B. Yuan, A. Ahmad, B. A. Shah, G. Mehboob, W. Lin, S. Ajmal, W. Peng, G. Yang, J. Liu and R. Hu, Direct Regeneration of Highly Degraded Cathode Materials from Spent Lithium-Ion Batteries *via* Eutectic Molten Salts with CoO and MnO2 Additives, *Chem. Eng. J.*, 2025, **517**, 164257, DOI: 10.1016/j.cej.2025.164257.

48 Z. Huang, D. Yu, B. Makuza, Q. Tian, X. Guo and K. Zhang, Hydrogen Reduction of Spent Lithium-Ion Battery Cathode Material for Metal Recovery: Mechanism and Kinetics, *Front. Chem.*, 2022, **10**, 1019493, DOI: 10.3389/fchem.2022.1019493.

49 Y. Shi, M. Zhang, Y. S. Meng and Z. Chen, Ambient-Pressure Relithiation of Degraded LiNi0.5Co0.2Mn0.3O2 $(0 < x < 1)$ *via* Eutectic Solutions for Direct Regeneration of Lithium-Ion Battery Cathodes, *Adv. Energy Mater.*, 2019, **9**(20), 1900454, DOI: 10.1002/aenm.201900454.

50 T. Wang, H. Luo, Y. Bai, I. Belharouak, K. Jayanthi, M. P. Paranthaman, B. T. Manard, E. T.-H. Wang, F. Dogan, S.-B. Son, B. J. Ingram, Q. Dai and S. Dai, Direct Recycling of Spent Nickel-Rich Cathodes in Reciprocal Ternary Molten Salts, *J. Power Sources*, 2024, **593**, 233798, DOI: 10.1016/j.jpowsour.2023.233798.

51 P. Parikh, H. Chung, E. Vo, A. Banerjee, Y. S. Meng and A. Devaraj, Nanoscale Compositional Mapping of Commercial LiNi0.8Co0.15Al0.05O2 Cathodes Using Atom Probe Tomography, *J. Phys. Chem. C*, 2022, **126**(34), 14380–14388, DOI: 10.1021/acs.jpcc.2c01217.

52 J. Li, W. Zhong, Q. Deng, Q. Zhang and C. Yang, Recent Progress in Synthesis and Surface Modification of Nickel-Rich Layered Oxide Cathode Materials for Lithium-Ion Batteries, *Int. J. Extrem. Manuf.*, 2022, **4**(4), 042004, DOI: 10.1088/2631-7990/ac92ef.

53 B. Jin, Z. Cui and A. Manthiram, In Situ Interweaved Binder Framework Mitigating the Structural and Interphasial Degradations of High-Nickel Cathodes in Lithium-Ion Batteries, *Angew. Chem., Int. Ed.*, 2023, **62**(15), e202301241, DOI: 10.1002/anie.202301241.

54 J. Song, L. Zhu, Y. Li, E. Han, Q. Zhang, G. Chen, Z. Zhang, X. Yang and Y. W. He, Modification of Nickel-Rich Ternary Cathode Material for Efficient Lithium-Ion Batteries, *J. Electrochem. Soc.*, 2023, **170**(1), 010523, DOI: 10.1149/1945-7111/acb0b9.

55 Y. Jiang, F. Guo, L. Qiu, T. Liu, Y. Hu, W. Yang, Y. Liu, Y. Sun, Z. Wu, Y. Song and X. Guo, Enhancing the Integral Structural and Thermal Stability of Ultrahigh-Ni Cathodes *via* Morphology Refinement and *In Situ* Interfacial Engineering, *ACS Appl. Mater. Interfaces*, 2023, **15**(29), 35072–35081, DOI: 10.1021/acsami.3c07022.

56 Y. Xiao, J. Su and L. Chen, A Comparative Analysis of Cathode Stripping Methods for Direct Recycling of Spent Li-Ion Batteries, *Proc. ASME 2023 18th Int. Manuf. Sci. Eng. Conf.*, 2023, vol. 2, p. V002T06A036, DOI: 10.1115/MSEC2023-105595.

57 J. Chacana-Olivares, B. Peceño, M. Grageda, C. Cruz and L. Rojas, Lithium-Ion Battery Recycling: A Perspective on Key Challenges and Opportunities, *npj Mater. Sustainability*, 2025, **3**(1), 38, DOI: 10.1038/s44296-025-00083-7.

58 M. Łach, A. Przybek, P. Duda and P. Bielaczyc, A Review of Direct Recycling Processes for Lithium-Ion Battery Cells, *Materials*, 2025, **18**(24), 5608, DOI: 10.3390/ma18245608.

59 J. Lee and J. Lopez, A Rapid Low-Temperature Method for Direct Recycling of Lithium-Ion Battery Cathodes, *Meet. Abstr.*, 2024, (55), 2948, DOI: 10.1149/MA2024-01552948mtgabs.

60 Y. Zhou, Z. Chen, A. Chen, J. Zhang, X. Wu and J. Xu, Comprehensive Recovery of NCM Cathode Materials for Spent Lithium-Ion Batteries by Microfluidic Device, *Sep. Purif. Technol.*, 2022, **294**, 121185, DOI: 10.1016/j.seppur.2022.121185.

61 T. Liu, J. Chen, X. Shen and H. Li, Regulating and Regenerating the Valuable Metals from the Cathode Materials in Lithium-Ion Batteries by Nickel-Cobalt-Manganese Co-Extraction, *Sep. Purif. Technol.*, 2021, **259**, 118088, DOI: 10.1016/j.seppur.2020.118088.

62 J. C.-Y. Jung, N. Chow, A. Nacu, M. Melashvili, A. Cao, L. Khorbaladze, Z. Meseldzija, J. P.-C. Sui and J. Zhang, A novel closed loop process for recycling spent li-ion battery cathode materials, *Int. J. Green Energy*, 2021, **18**(15), 1597–1612, DOI: 10.1080/15435075.2021.1914631.

63 T. Yun, J. Kim, S. Lee and S. Hong, Application of Vacuum Membrane Distillation Process for Lithium Recovery in Spent Lithium Ion Batteries (LIBs) Recycling Process, *Desalination*, 2023, **565**, 116874, DOI: 10.1016/j.desal.2023.116874.

64 G. Qian, Z. Li, Y. Wang, X. Xie, Y. He, J. Li, Y. Zhu, S. Xie, Z. Cheng, H. Che, Y. Shen, L. Chen, X. Huang, P. Pianetta, Z.-F. Ma, Y. Liu and L. Li, Value-Creating Upcycling of Retired Electric Vehicle Battery Cathodes, *Cell Rep. Phys. Sci.*, 2022, **3**(2), 100741, DOI: 10.1016/j.xcrp.2022.100741.

65 X. Ye, X. Fei, M. Liu, H. Gao, B. Qiu, H. Yin, Z. Zhang and L. Y. S. Lee, Laser-Induced Regeneration of Spent LiMn2O4 Cathode Into High-Performance Ni-Doped LiMn2O4 Cathode, *Adv. Mater.*, 2025, **37**(5), 2416537, DOI: 10.1002/adma.202416537.

66 S. Gu, J. Kong, L. Xing, X. Zhu, J. Xu, C. Chen and Z. Zhang, Feasible Regeneration of Cathode Material from Spent Portable Electronics Batteries *via* Nano-Bubbles Enhanced

Open Access Article. Published on 18 May 2026. Downloaded on 5/21/2026 12:16:01 PM.
This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence.

Leaching, *J. Clean. Prod.*, 2022, **368**, 133199, DOI: 10.1016/j.jclepro.2022.133199.

67 L. Schlott, M. Gutsch and J. Leker, Cost Modelling and Key Drivers in Lithium-Ion Battery Recycling, *Nat. Rev. Clean Technol.*, 2025, **1**(9), 656–670, DOI: 10.1038/s44359-025-00095-5.

68 Q. Hoarau and E. Lorang, An Assessment of the European Regulation on Battery Recycling for Electric Vehicles, *Energy Policy*, 2022, **162**, 112770, DOI: 10.1016/j.enpol.2021.112770.

69 Y. Zong, P. Yao, X. Zhang, J. Wang, X. Song, J. Zhao, Z. Wang and Y. Zheng, Material Flow Analysis on the Critical Resources from Spent Power Lithium-Ion Batteries under the Framework of China's Recycling Policies, *Waste Manage.*, 2023, **171**, 463–472, DOI: 10.1016/j.wasman.2023.09.039.

70 Q. Zhao, K. Sun, X. Wang, Q. Wang and J. Wang, Examining Green-Sustainable Approaches for Recycling of Lithium-Ion Batteries, *Carbon*, 2024, **3**, 100034, DOI: 10.1016/j.decarb.2023.100034.

71 A. Pražanová, Z. Plachý, J. Kočí, M. Fridrich and V. Knap, Direct Recycling Technology for Spent Lithium-Ion Batteries: Limitations of Current Implementation, *Batteries*, 2024, **10**(3), 81, DOI: 10.3390/batteries10030081.

72 H. Zhou, W. Li, T. Poulet, H. Basarir and A. Karrech, Life Cycle Assessment of Recycling Lithium-Ion Battery Related Mineral Processing by-Products: A Review, *Miner. Eng.*, 2024, **208**, 108600, DOI: 10.1016/j.mineng.2024.108600.

73 Y. Lu, X. Han and Z. Li, Enabling Intelligent Recovery of Critical Materials from Li-Ion Battery through Direct Recycling Process with Internet-of-Things, *Materials*, 2021, **14**(23), 7153, DOI: 10.3390/ma14237153.

74 L. L. Driscoll, A. Jarvis, R. Madge, E. H. Driscoll, J.-M. Price, R. Sommerville, F. S. Tontini, M. Bahri, M. Miah, B. L. Mehdi, E. Kendrick, N. D. Browning, P. K. Allan, P. A. Anderson and P. R. Slater, Phase-Selective Recovery and Regeneration of End-of-Life Electric Vehicle Blended Cathodes *via* Selective Leaching and Direct Recycling, *Joule*, 2024, **8**(10), 2735–2754, DOI: 10.1016/j.joule.2024.07.001.

75 Z.-H. Dai, J. Wang, H. Zhao and Y. Bai, Surface Coupling between Mechanical and Electric Fields Empowering Ni-Rich Cathodes with Superior Cyclabilities for Lithium-Ion Batteries, *Adv. Sci.*, 2022, **9**, 2200622, DOI: 10.1002/advs.202200622.

76 D. Gastol, J. Marshall, E. Cooper, C. Mitchell, D. Burnett, T. Song, R. Sommerville, B. Middleton, M. Crozier, R. Smith, S. Haig, C. R. McElroy, N. van Dijk, P. Croft, V. Goodship and E. Kendrick, Reclaimed and Up-Cycled Cathodes for Lithium-Ion Batteries, *Glob. Chall.*, 2022, **6**(12), 2200046, DOI: 10.1002/gch2.202200046.

77 G. Ji, D. Tang, J. Wang, Z. Liang, H. Ji, J. Ma, Z. Zhuang, S. Liu, G. Zhou and H.-M. Cheng, Sustainable Upcycling of Mixed Spent Cathodes to a High-Voltage Polyanionic Cathode Material, *Nat. Commun.*, 2024, **15**(1), 4086, DOI: 10.1038/s41467-024-48181-9.

78 F. Wang, Y. Deng and C. Yuan, Life Cycle Assessment of Lithium Oxygen Battery for Electric Vehicles, *J. Clean. Prod.*, 2020, **264**, 121339, DOI: 10.1016/j.jclepro.2020.121339.

79 M. Hiremath, K. Derendorf and T. Vogt, Comparative Life Cycle Assessment of Battery Storage Systems for Stationary Applications, *Environ. Sci. Technol.*, 2015, **49**(8), 4825–4833, DOI: 10.1021/es504572q.

80 G. Patrizi, F. Canzanella, L. Ciani and M. Catelani, Life Cycle Analysis of Lithium Batteries for Smart Grids Using Electrochemical Impedance Spectroscopy Data, in *2024 IEEE International Workshop on Metrology for Industry 4.0 & IoT (MetroInd4.0 & IoT)*, 2024, pp 298–303, DOI: 10.1109/MetroInd4.0IoT61288.2024.10584119.

81 C. Xing, M. Yao and L. Fei, Upcycling Degraded Layered Oxide Cathodes from Spent Lithium-Ion Batteries toward Emerging Materials: A Review, *Energy Storage Mater.*, 2024, **71**, 103636, DOI: 10.1016/j.ensm.2024.103636.

82 L. Sugiarto and Y.-C. Lu, Accurate Battery Life Prediction Using Early-Cycle Features, *Meet. Abstr.*, 2024, (53), 2800, DOI: 10.1149/MA2024-01532800mtgabs.

83 H. Yin, J. Xiao and G. Wang, Human-Robot Collaboration Re-Manufacturing for Uncertain Disassembly in Retired Battery Recycling, in *2022 5th World Conference on Mechanical Engineering and Intelligent Manufacturing (WCMEIM)*, 2022; pp pp 595–598, DOI: 10.1109/WCMEIM56910.2022.10021388.

84 X. Ma, Z. Meng, M. V. Bellonia, J. Spangenberger, G. Harper, E. Gratz, E. Olivetti, R. Arsenault and Y. Wang, The Evolution of Lithium-Ion Battery Recycling, *Nat. Rev. Clean Technol.*, 2025, **1**(1), 75–94, DOI: 10.1038/s44359-024-00010-4.

85 Umicore Battery Recycling. https://www.umicore.com/en/media/newsroom/umicore-battery-recycling/(accessed 2025-04-21).

86 S. Antony Jose, C. A. D. Cook, J. Palacios, H. Seo, C. E. Torres Ramirez, J. Wu and P. L. Menezes, Recent Advancements in Artificial Intelligence in Battery Recycling, *Batteries*, 2024, **10**(12), 440, DOI: 10.3390/batteries10120440.

87 Y. Hua, S. Zhou, Y. Huang, X. Liu, H. Ling, X. Zhou, C. Zhang and S. Yang, Sustainable Value Chain of Retired Lithium-Ion Batteries for Electric Vehicles, *J. Power Sources*, 2020, **478**, 228753, DOI: 10.1016/j.jpowsour.2020.228753.

© 2026 The Author(s). Published by the Royal Society of Chemistry