Direct Recycling of Lithium-Ion Batteries: A Survey on Failure Mechanisms, Surface Reconstruction, and Interface Modification for Enhanced Sustainability

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Abstract

This survey paper examines the role of direct recycling methods in enhancing the sustainability of lithium-ion batteries (LIBs) by addressing failure mechanisms and optimizing material recovery processes. The analysis highlights the importance of direct recycling techniques such as Molten-Salt Direct-Recycling (MSDR), which efficiently regenerate high-performance cathodes and support closed-loop recycling initiatives. These methods not only conserve valuable resources but also reduce the environmental footprint associated with battery production and disposal. The survey underscores the necessity of developing efficient recycling processes to improve recovery rates of critical materials like lithium, essential for the economic viability and sustainability of LIB technologies. Furthermore, it emphasizes the need to address safety issues through improved materials and design, as LIBs are pivotal to sustainable energy solutions. Advanced analytical techniques and modeling frameworks are crucial for optimizing recycling processes, providing insights into the structural and electrochemical properties of LIB materials. These innovations facilitate the development of more effective recycling strategies, contributing to the sustainable growth of energy storage technologies. By leveraging these advancements, the direct recycling of LIBs can be conducted more efficiently and sustainably, aligning with broader goals of environmental protection and resource conservation.

1 Introduction

1.1 Importance of Lithium-Ion Battery Recycling

The recycling of lithium-ion batteries (LIBs) is driven by pressing environmental and economic considerations. Environmentally, improper disposal of LIBs generates hazardous waste, necessitating stable electrolytes to enhance performance and mitigate impacts [1]. Accurate characterization of LIB internal states is essential for performance, durability, and safety [2]. Additionally, rapid capacity degradation in certain anode materials, such as SnO-based anodes, highlights the urgent need for improved recycling methods to manage waste and recover valuable materials [3].

Economically, the electrification of the automotive industry has spurred a surge in battery production, creating challenges in sourcing key materials and recycling retired batteries [4]. Recycling is critical for recovering scarce materials, reducing dependency on primary resources, and stabilizing supply chains [5]. The demand for enhanced reliability, safety, and performance in energy storage technologies further underscores the necessity of recycling [6]. Moreover, the limitations of traditional graphite anodes necessitate the development of high-performance negative electrode materials, reinforcing the economic case for recycling to boost battery performance and sustainability [7].

Recycling also addresses safety concerns, particularly the thermal runaway risks associated with LIBs in electric vehicles [8]. By integrating sustainable practices in battery management, recycling extends

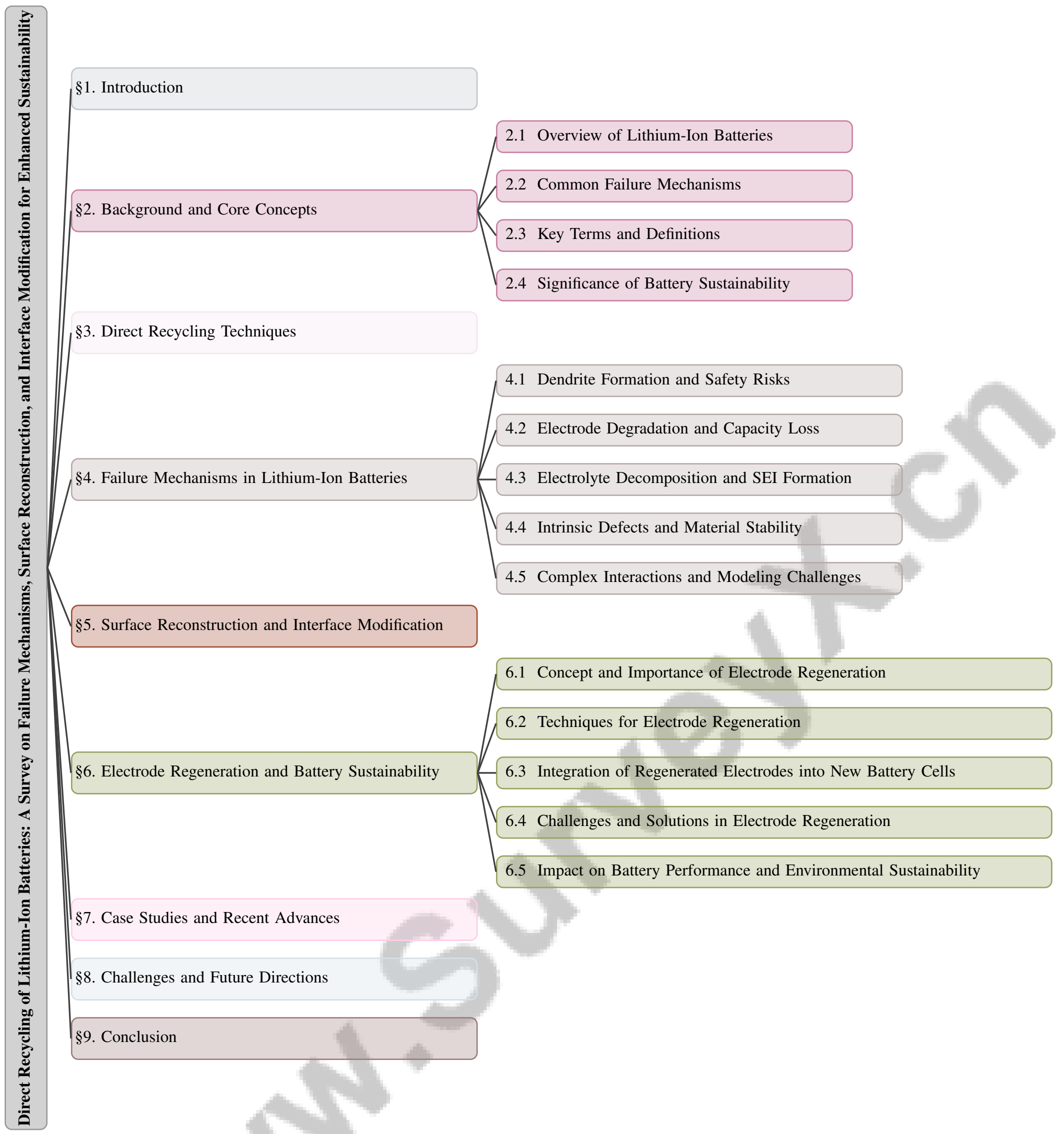

Figure 1: chapter structure

LIB lifespans and promotes sustainable utilization. Addressing both environmental and economic challenges, recycling is a pivotal mechanism for the long-term sustainability of lithium-ion batteries.

1.2 Focus on Direct Recycling Methods

Direct recycling methods are crucial for enhancing the sustainability and performance of lithium-ion batteries (LIBs) by addressing critical failure mechanisms and optimizing material recovery processes. This survey emphasizes these methods, which rejuvenate battery components directly without extensive chemical processing, thereby preserving the intrinsic properties of battery materials. The evolution of materials and technologies facilitating LIB commercialization underscores the significance of direct recycling in advancing battery sustainability [1].

Innovative techniques such as phosphorus doping enhance the electrochemical performance of LIBs through the creation of protrusions in graphene [9]. Complementary non-invasive ultrasound technology allows for battery health evaluation without disassembly, maintaining cell integrity for reuse [2]. Such advancements are critical for mitigating safety risks like thermal runaway and battery fires, which are paramount concerns in direct recycling contexts [10].

Molten-salt direct-recycling (MSDR) techniques exemplify the environmental and economic viability of direct recycling, enabling the upcycling of low-nickel polycrystalline cathodes into nickel-rich single-crystal cathodes, enhancing both material value and performance [11]. The adaptability of direct recycling methods extends to batteries rich in cobalt, nickel, and manganese, showcasing their versatility across various battery chemistries [12].

Furthermore, the direct regeneration of spent LiFePO4 cathodes using multifunctional organic lithium salts illustrates the effectiveness of direct recycling in restoring battery components [13]. Incorporating computational techniques, such as those for computing Madelung energies in ionic crystals, can optimize material recovery and enhance battery performance [14].

This survey highlights the critical role of direct recycling in advancing LIB sustainability through innovative technologies and methodologies that address performance and safety challenges. Such efforts significantly contribute to the long-term viability of battery technologies within sustainable energy systems [15]. In evaluating environmental impacts and economic implications, direct recycling methods emerge as a focal point in broader LIB recycling strategies [4].

1.3 Structure of the Survey

This survey is organized into several key sections, each exploring different aspects of lithium-ion battery (LIB) recycling, with a specific focus on direct recycling methods. The introduction emphasizes the importance of sustainable recycling practices for LIBs, setting the stage for the in-depth analysis that follows.

The first section provides background information on core concepts, offering an overview of LIBs, their components, and common failure mechanisms. It defines key terms such as direct recycling, surface reconstruction, and interface modification, underscoring the significance of battery sustainability.

The subsequent section delves into direct recycling techniques, examining various methods and their advantages over traditional approaches. This part includes a discussion on molten-salt direct-recycling (MSDR) and contrasts direct recycling methods with conventional hydrometallurgical techniques, as outlined in the framework categorizing LIB recycling methods [12]. It also explores hybrid models, recent innovations, and the economic and environmental implications of these methods.

The survey addresses various failure mechanisms affecting LIBs, focusing on critical issues such as dendrite formation, which can lead to short circuits, electrode degradation impacting overall capacity and performance, and electrolyte decomposition compromising battery stability and safety. Utilizing advanced degradation mode analysis, the study elucidates complex interactions between these mechanisms and their contributions to capacity and power fade, thereby enhancing our understanding of battery longevity and informing future design improvements [16, 17, 18]. It also addresses intrinsic defects and challenges in modeling complex interactions within battery systems.

The focus then shifts to surface reconstruction and interface modification processes, detailing techniques like phosphorus doping and the use of graphene protrusions. Advanced imaging and modeling techniques, along with the states-filling model for surface reconstruction, are discussed. This section covers atomic layer deposition for interface modification and grain boundary engineering with solid electrolyte infusion.

The survey continues with a section on electrode regeneration, highlighting its role in enhancing battery sustainability. It discusses various techniques for regenerating electrodes and their integration into new battery cells, addressing challenges and solutions in this area.

The following section presents comprehensive case studies and recent advancements in direct recycling of LIBs, emphasizing innovative techniques and successful implementations that address the increasing demand for sustainable recycling solutions in light of the exponential growth in LIB usage, particularly in the electric transportation sector. These developments focus on overcoming limitations of traditional pyrometallurgical methods while recovering valuable materials and minimizing environmental impacts [19, 12]. This includes discussions on innovative analytical techniques and advancements in understanding phase transitions, as well as the impact of nanoscale insights on recycling practices.

The penultimate section identifies challenges and future directions in direct recycling, exploring technological limitations, economic feasibility, regulatory and safety challenges, and material and

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design innovations. It examines the integration of advanced technologies and potential future research opportunities.

Finally, the survey concludes by summarizing key findings, emphasizing the importance of direct recycling in addressing failure mechanisms and promoting LIB sustainability. This comprehensive structure provides a framework for understanding the lifecycle of LIBs, with a strong emphasis on recycling and environmental sustainability [15].The following sections are organized as shown in Figure 1.

2 Background and Core Concepts

2.1 Overview of Lithium-Ion Batteries

Lithium-ion batteries (LIBs) are pivotal in energy storage due to their high energy density and efficiency, crucial for carbon neutrality technologies in electric vehicles and portable electronics [20, 2]. Key components include the anode, often graphite or lithium metal, which stores lithium ions during charging, and the cathode, typically metal oxides or chalcogenides, facilitating ion movement during discharge [1]. Early electric vehicles used low-nickel LiNi0.5Mn0.3Co0.2O2 cathodes, highlighting the need for efficient recycling [11].

The electrolyte, a lithium salt in an organic solvent, ensures ionic conductivity, while the separator prevents direct contact between electrodes. Structural enhancements, like graphene, improve mechanical and electrochemical stability [21]. Innovations in materials, such as Ga-In-Sn alloy with PVDF and NMP, further advance LIB performance and sustainability [7]. These components collectively ensure LIBs' functionality and efficiency, establishing them as essential in modern energy storage.

2.2 Common Failure Mechanisms

LIBs are susceptible to failure mechanisms affecting performance, safety, and lifespan. Dendrite formation in lithium metal anodes can cause internal short circuits, increasing thermal runaway risks [5, 22]. This growth is exacerbated by uneven current density, promoting localized failure. Electrode degradation, especially in silicon-based electrodes, results from volume changes during lithiation, causing cracks [23]. Diffusion-induced stresses in core-shell structures further compound this issue [24]. Intrinsic defects in lithium manganese oxides also limit performance [25].

Electrolyte decomposition, particularly ethylene carbonate breakdown, is critical during SEI formation, which is essential yet can lead to lithium-ion depletion if unstable [26]. Liquid electrolyte penetration into grain boundaries exacerbates intergranular cracking, causing instability [27]. Current models inadequately represent ion transport and intercalation, often overlooking critical factors like particle size and porosity, complicating degradation prediction [28]. The interplay of degradation mechanisms necessitates advanced recycling and modeling techniques for material recovery and environmental impact mitigation. Comprehensive degradation analysis is crucial for lifespan prediction and recycling optimization, requiring innovative modeling approaches to manage LIB end-of-life pathways [16, 15, 18, 19].

2.3 Key Terms and Definitions

Understanding key terms is crucial for navigating lithium-ion battery recycling and performance optimization, especially with rising demand from electric vehicles and environmental challenges [15, 4]. Direct recycling restores battery components without extensive chemical alteration, preserving material properties, unlike traditional methods involving complete breakdown and synthesis.

Surface reconstruction enhances electrode surfaces using techniques like phosphorus doping and graphene incorporation, improving lithium adsorption and conductivity [9]. These methods address issues in carbon-based anodes' performance [29]. Interface modification enhances ionic transport and reduces degradation, with the SEI acting as a barrier for lithium-ion transport while preventing further decomposition [30]. Boron nitride flakes serve as protective layers, enhancing stability and safety [8].

Electrode regeneration restores or improves performance through reversible lithiation techniques, crucial for capacity and longevity, as shown in the SnO lithiation model [3]. Understanding EC decomposition pathways is vital, influencing SEI formation energetics and kinetics [31].

These terms provide a framework for addressing challenges and innovations in LIB recycling and performance enhancement, crucial for mitigating environmental impacts from production and disposal. Evaluating recycling methods and technological advancements is essential for developing sustainable energy storage solutions, aiding resource recovery and reducing ecological harm [15, 19, 4, 12, 18].

2.4 Significance of Battery Sustainability

The sustainability of LIBs is critical in energy storage, driven by increasing demand in electronics, vehicles, and renewable systems [1]. The environmental impact from material extraction and production pollution necessitates sustainable battery practices [4]. Efficient recycling is essential for recovering valuable metals like lithium and mitigating disposal impacts [5].

Safety is pivotal for sustainability; predicting LIBs' Remaining Useful Life (RUL) is crucial for reliability [6]. Advanced modeling frameworks improve degradation interaction understanding, supporting sustainable management [18]. Developing materials that balance energy density with safety is vital for sustainable technology [5].

Non-destructive techniques, like magnetic imaging, visualize conductivity changes, enhancing understanding and supporting sustainability [32]. Methods preventing intergranular cracking and enhancing cycle stability, such as tailored grain boundary structures, contribute to LIB longevity [27]. Accurate ion transport representation, considering particle size and transport percolation, provides insights for optimizing design and sustainability [28]. A comprehensive understanding of LIBs is essential for efficient use in vehicles and renewable systems, contributing to sustainable energy solutions [26].

Efforts focus on minimizing environmental impacts from LIB production and disposal, enhancing longevity through improved degradation models and recycling methods, and promoting safe, efficient use in various applications, particularly in the growing electric vehicle market. These initiatives address critical issues in material extraction and recycling, ensuring a sustainable supply chain and reducing waste management challenges as demand rises [5, 15, 4, 12, 18]. Integrating recycling processes and innovative material designs underscores sustainability's importance in battery technology, promoting a sustainable future for energy storage systems.

In recent years, the focus on sustainable practices in battery recycling has intensified, particularly in the context of lithium-ion batteries. The exploration of direct recycling techniques has emerged as a pivotal area of research. Figure 2 illustrates the hierarchical structure of these direct recycling techniques, emphasizing the innovative Molten-Salt Direct-Recycling (MSDR) process. This figure not only compares MSDR with traditional recycling methods but also showcases the integration of hybrid models and innovations. Furthermore, it highlights the economic and environmental implications of these advanced recycling strategies, thereby providing a comprehensive overview of the current landscape in battery recycling technologies. This visual representation serves to enhance our understanding of the complexities involved in the recycling processes and their potential impact on sustainability efforts.

3 Direct Recycling Techniques

3.1 Molten-Salt Direct-Recycling (MSDR)

Molten-Salt Direct-Recycling (MSDR) is an innovative technique for recycling lithium-ion batteries (LIBs) that focuses on regenerating high-value cathode materials with minimal chemical modification [11]. Utilizing the unique properties of molten salts, MSDR selectively recovers and transforms cathode materials, preserving their crystal structure and enhancing electrochemical performance [12]. This process involves dissolving spent cathode materials in a molten salt medium, efficiently recovering valuable metals while maintaining the crystal integrity, enabling the production of nickel-rich single-crystal cathodes from low-nickel polycrystalline sources [11]. By regenerating high-performance cathodes without extensive re-synthesis, MSDR reduces environmental impact and energy consumption associated with traditional recycling methods [19].

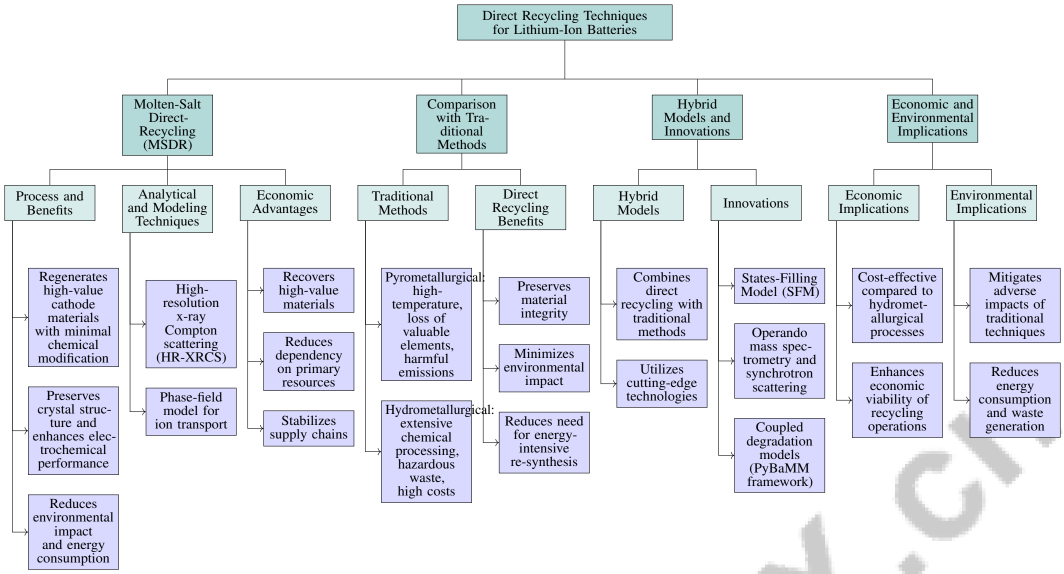

Figure 2: This figure illustrates the hierarchical structure of direct recycling techniques for lithium-ion batteries, highlighting the innovative Molten-Salt Direct-Recycling (MSDR) process, its comparison with traditional methods, the integration of hybrid models and innovations, and the economic and environmental implications of these advanced recycling strategies.

As illustrated in Figure 3, the figure provides a comprehensive overview of the MSDR technique, showcasing its structural components and the various technological benefits it offers. Additionally, it highlights the advanced analytical techniques involved, demonstrating the method's potential to enhance lithium-ion battery recycling. Advanced analytical techniques, such as high-resolution x-ray Compton scattering (HR-XRCS), provide insights into the redox orbitals and electron momentum density during lithium insertion and extraction, optimizing recycling processes and enhancing battery performance [33]. MSDR's compatibility with innovative modeling frameworks, like the phase-field model for ion transport, which accounts for particle size distribution and percolative ion transport in porous electrodes, improves the efficiency of direct recycling methods [28]. Economically, MSDR offers significant advantages by recovering high-value materials, reducing dependency on primary resources, and stabilizing supply chains [19]. As global demand for LIBs escalates, the development and implementation of efficient recycling techniques like MSDR are crucial for supporting sustainable growth in the energy storage sector [12].

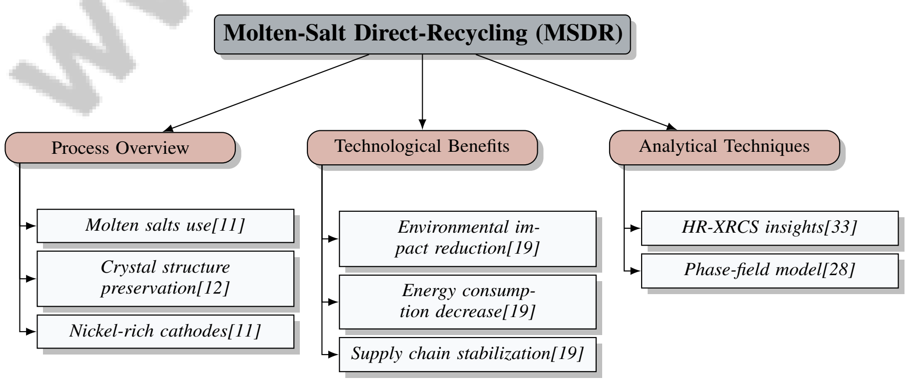

Figure 3: This figure illustrates the structure and advantages of the Molten-Salt Direct-Recycling (MSDR) technique. It highlights the process overview, technological benefits, and analytical techniques involved, demonstrating the method's potential to enhance lithium-ion battery recycling.

3.2 Comparison with Traditional Methods

Direct recycling methods for LIBs provide significant benefits over traditional pyrometallurgical and hydrometallurgical approaches by preserving material integrity and minimizing environmental impact [12]. Traditional pyrometallurgical techniques, which involve high-temperature processes to melt battery components, often result in the loss of valuable elements and harmful gas emissions [11], while yielding low recovery efficiencies for critical materials like lithium [19]. Hydrometallurgical methods, although capable of recovering a broader range of materials, involve extensive chemical processing, generating hazardous waste and increasing operational costs [4]. The chemical dissolution in hydrometallurgical recycling can degrade the cathode materials' crystal structure, necessitating complete re-synthesis [12].

In contrast, direct recycling methods such as MSDR focus on the selective recovery and regeneration of cathode materials with minimal chemical alteration, preserving their crystal structure and enhancing electrochemical performance [11]. This approach reduces the need for energy-intensive re-synthesis processes, aligning with environmental sustainability goals by minimizing waste generation and energy consumption [19]. Advanced analytical and modeling tools, such as HR-XRCS and phase-field models for ion transport, optimize material recovery processes by providing insights into the structural and electrochemical properties of recycled battery materials [34, 33, 11]. The shift towards direct recycling methods signifies a substantial advancement in sustainable LIB management, offering a more environmentally friendly and economically viable alternative to traditional practices. As LIB demand surges, driven by electric vehicles and advancements in energy storage technologies, effective direct recycling techniques will be essential for mitigating environmental impacts and ensuring a sustainable supply of critical materials [15, 19, 12].

3.3 Hybrid Models and Innovations

Hybrid recycling models represent a significant advancement in LIB recycling by merging various techniques to optimize material recovery and enhance sustainability. These models combine direct recycling techniques with traditional methodologies, utilizing cutting-edge technologies to address challenges posed by diverse LIB compositions, thereby improving material recovery efficiency and minimizing environmental impact [12, 15, 4, 19]. A notable innovation is the incorporation of the States-Filling Model (SFM), which relates the binding energy of lithium on sp2 carbon substrates to the work required to fill previously unoccupied electronic states, enhancing understanding of electronic interactions during recycling processes [29].

Recent advancements focus on integrating analytical techniques, such as operando mass spectrometry and synchrotron scattering, to map reaction mechanisms during battery operation, offering insights into degradation processes and facilitating more effective recycling strategies [35]. Coupled degradation models, such as those within the PyBaMM framework, integrate multiple degradation mechanisms—SEI growth, lithium plating, particle cracking, and loss of active material—providing a comprehensive understanding of battery degradation and informing efficient recycling practices [16]. Hybrid models also explore the infusion of protective materials into battery components to prevent degradation, such as infusing Li3PO4 into grain boundaries to prevent liquid electrolyte penetration and mitigate side reactions [27]. These innovations demonstrate the potential for combining advanced analytical techniques, theoretical models, and protective material infusions to optimize LIB recycling, enhancing the accuracy of lifetime predictions and contributing to more sustainable energy storage solutions [16, 15, 18].

3.4 Economic and Environmental Implications

The economic and environmental implications of direct recycling methods, particularly MSDR, present significant advantages over traditional recycling processes. MSDR is cost-effective compared to hydrometallurgical processes, reducing processing expenses while minimizing environmental impact. This method facilitates the direct production of high-value cathodes from retired battery materials, enhancing the economic viability of recycling operations [11]. By regenerating valuable cathode materials without extensive chemical reprocessing, operational costs are reduced, and supply chains are stabilized by decreasing reliance on primary resources. This is crucial given the rising demand for LIBs driven by transportation electrification and the expansion of renewable energy systems [15].

Advanced technologies, such as ParaSweeper, which can perform a million charge/discharge cycles daily, accelerate the exploration of battery degradation models, improving battery design and longevity [36]. Environmentally, direct recycling methods mitigate adverse impacts associated with traditional techniques by preserving the crystal structure and intrinsic properties of battery materials, reducing energy consumption and waste generation typical of pyrometallurgical and hydrometallurgical processes [11]. This reduced environmental footprint aligns with sustainable practices to mitigate the impacts of battery production and disposal on ecosystems [15]. Advanced monitoring techniques, such as the Gaussian Process-based Electrochemical Model (GP-ECM), enhance the safety and reliability of recycling processes by providing real-time monitoring capabilities and early fault detection [37], ensuring optimal recycling conditions and minimizing environmental contamination, thus improving the sustainability of battery recycling practices.

4 Failure Mechanisms in Lithium-Ion Batteries

Understanding lithium-ion batteries (LIBs) involves examining specific failure mechanisms that impact safety and performance. Among these, dendrite formation is particularly critical, affecting battery efficiency and safety. The following subsection delves into dendrite formation processes and their implications for LIB reliability and safety.

4.1 Dendrite Formation and Safety Risks

Dendrite formation in LIBs is a significant safety and performance concern. These needle-like lithium structures can grow during charging, especially in lithium metal anodes, leading to internal short circuits, thermal runaway, and fires [17]. High-resolution imaging from CEPRI has revealed insights into these structures, highlighting current distribution and safety hazards [17]. This phenomenon is linked to lithium plating, a critical factor affecting both performance and safety [38]. Uneven current distribution exacerbates dendrite growth, resulting in large voltage hysteresis, as observed in FeF3 electrodes, compromising performance [39]. Mechanical failures, such as fracture and debonding in core-shell structures, further increase safety risks, underscoring the need for robust mechanical designs to prevent degradation [24].

Gas-induced bulging, where gas formation deforms pouch cells, also poses a safety risk [40]. Mitigation strategies, such as the interaction of boron nitride flakes with lithium atoms, aim to stabilize the electrode surface and reduce dendritic growth [8]. The reversible changes in thermal conductivity and elastic modulus in intercalation materials, contrasted with irreversible losses in conversion materials, highlight the complex interplay between material properties and dendrite formation [41].

Figure 4 illustrates the hierarchical structure of key issues and mitigation strategies related to dendrite formation and safety risks in lithium-ion batteries. This figure highlights the primary concerns of dendrite growth, safety concerns, and proposed mitigation strategies, providing a visual representation that complements the discussion on the critical nature of addressing dendrite formation. Addressing these challenges is crucial for developing safer, more reliable energy storage solutions as the demand for high-performance batteries continues to rise.

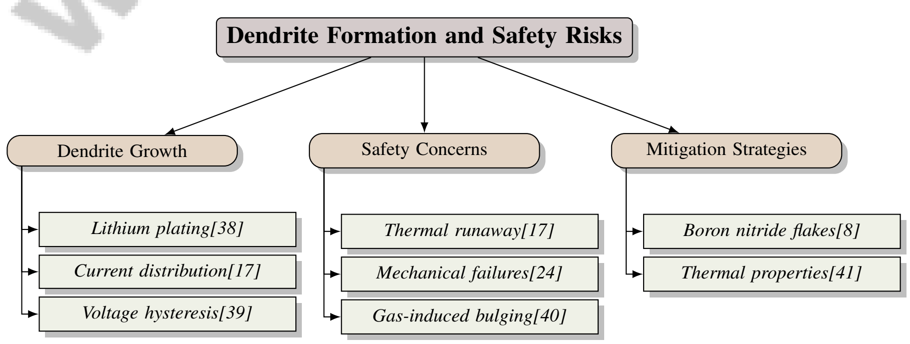

Figure 4: This figure illustrates the hierarchical structure of key issues and mitigation strategies related to dendrite formation and safety risks in lithium-ion batteries. It highlights the primary concerns of dendrite growth, safety concerns, and proposed mitigation strategies.

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4.2 Electrode Degradation and Capacity Loss

Electrode degradation in LIBs is a complex phenomenon significantly affecting capacity and performance. Mechanical and chemical stresses during cycling drive this degradation. For example, lithiation in silicon-based anodes induces substantial volume changes, leading to plastic deformation at stresses up to 1.48 GPa, with fracture energies estimated between 9-11 J/m^2 [23]. Such stresses result in irreversible damage, impairing the anode's ability to accommodate lithium ions effectively.

In graphene electrodes, mechanical strain exacerbates degradation, leading to capacity loss during lithium intercalation [21]. This strain-induced degradation illustrates the intricate relationship between mechanical integrity and electrochemical performance, where stresses enhancing stability may detrimentally impact electrochemical properties. Structural transformations in SnO anodes during lithiation critically influence degradation, impacting overall capacity and performance [3]. These transformations can lead to cracks and defects, exacerbating capacity loss and reducing battery lifespan.

The interplay between mechanical stresses and electrochemical performance is further complicated by stress evolution in composite electrodes. Real-time stress measurements using the wafer-curvature method have provided insights into how stress changes during operation contribute to degradation [23]. For example, germanium thin film electrodes exhibit significant plastic deformation during lithiation, highlighting the mechanical challenges faced by electrode materials.

The degradation of electrodes in LIBs significantly impacts capacity and sustainability, involving complex interactions among various mechanisms such as loss of lithium inventory and active material, leading to capacity fade and limiting effective lifespan. Recent advancements in degradation mode analysis emphasize the importance of understanding these interactions to improve lifetime predictions and optimize recycling processes, addressing performance and environmental concerns associated with LIBs [1, 10, 15, 16, 18]. Advanced modeling and estimation techniques are essential for accurately predicting degradation impacts on capacity, enhancing battery management systems, and extending LIB lifespan. Understanding and mitigating these processes are vital for improving energy storage technology efficiency and sustainability.

4.3 Electrolyte Decomposition and SEI Formation

Electrolyte decomposition and solid electrolyte interphase (SEI) formation are critical processes influencing the operation and longevity of LIBs. The SEI layer, forming on the anode surface during initial charging cycles, stabilizes the anode by preventing further electrolyte decomposition, thereby maintaining battery efficiency and lifespan [42]. SEI formation is complex, influenced by multiple reaction pathways contributing to its composition and properties.

The growth of the SEI is significantly affected by the availability of electrons in the reaction film, which restricts the rate of SEI formation. This electron-limited growth suggests a close relationship between SEI conductivity and lithium ion concentration within the layer [30]. As the SEI develops, it functions as a mixed ion-electron conductor (MIEC), facilitating lithium-ion transport while acting as a barrier to electron flow, crucial for maintaining electrochemical stability and mitigating capacity fade over time.

Electrolyte decomposition, particularly the reduction of ethylene carbonate (EC), is pivotal in SEI formation. The intricate pathways involved in SEI formation lead to a diverse array of organic and inorganic compounds that shape the SEI's structural integrity and functionality. Factors influencing growth dynamics include lithium intercalation, solvent and additive consumption during formation, and electrochemical kinetics governing capacity fade, all contributing to the SEI's effectiveness in enhancing battery performance and longevity [30, 31, 42]. The balance of these reactions determines the SEI's protective ability and overall battery performance.

Advanced modeling techniques have been employed to predict SEI formation, providing insights into the kinetics and thermodynamics of underlying reactions. These models elucidate the roles of different components in the SEI and guide strategies to enhance its formation and stability. By advancing our understanding of electrolyte decomposition and SEI growth mechanisms, researchers are developing sophisticated models that integrate electrochemical kinetics and degradation mode analysis to optimize LIB design and operational protocols. This comprehensive approach aims to enhance battery performance—by improving first-cycle efficiency and capacity retention—and prolong battery lifespan and sustainability by mitigating capacity fade and voltage instability through innovative strategies such as grain boundary engineering in cathode materials [30, 18, 27, 42].

4.4 Intrinsic Defects and Material Stability

Intrinsic defects in LIB materials significantly influence their stability and performance, playing a crucial role in degradation processes that affect battery lifespan. Defects such as vacancies, interstitials, and substitutions in the crystal lattice can alter electrochemical and mechanical properties. The presence of these defects can increase mechanical stresses and exacerbate degradation mechanisms, including loss of lithium inventory and active material, ultimately compromising battery stability and performance. This interplay of mechanical and electrochemical factors necessitates advanced degradation mode analysis for accurate lifetime predictions and improved design strategies for next-generation LIBs [16, 43, 18].

The phase-field model provides a robust framework for studying lithium-ion intercalation dynamics and associated mechanical stresses in battery particles, accounting for both exterior and interior interfaces [44]. This model emphasizes the significant role of intrinsic defects in influencing mechanical integrity, crucial for understanding microstructural evolution and electrode stability. The complex interplay between mechanical and electrochemical processes highlights the need for comprehensive modeling approaches, as demonstrated by the cohesive phase-field (CPF) interface model, which integrates cohesive zone and phase-field methods for accurate simulation of fracture patterns in LIB materials [45].

The impact of intrinsic defects is compounded by the coupling of multiple degradation mechanisms, as illustrated in models revealing exacerbated capacity fade at low temperatures due to active material loss [16]. These models underscore the need to consider interactions among various degradation pathways for accurate performance and longevity predictions.

Furthermore, a proposed framework for predicting void-driven damage in LIB electrodes offers insights into microstructural evolution, informing the design of next-generation electrodes with improved performance [34]. By capturing essential physics of charge transfer-dominated binding on planar carbon, rapid predictions of lithium binding characteristics based on electronic structure can be achieved, contributing to the development of more stable and efficient battery materials [29].

Addressing intrinsic defects in LIB materials is essential for enhancing stability and performance. Advanced modeling techniques, coupled with comprehensive analyses of interactions among multiple degradation mechanisms, provide critical insights into battery material design and optimization. This approach enhances understanding of complex degradation pathways—such as loss of lithium inventory and active material—and facilitates the development of more durable and reliable energy storage solutions. By integrating various degradation modes and utilizing advanced predictive models, researchers can better anticipate battery performance and lifespan under diverse operating conditions, ultimately improving safety, efficiency, and reducing warranty liabilities in energy storage applications [46, 16, 18].

4.5 Complex Interactions and Modeling Challenges

Modeling the complex interactions within LIBs presents significant challenges due to the intricate interplay between electrochemical reactions, mechanical behavior, and material stability. The phase-field model has emerged as a powerful tool for capturing these interactions, revealing insights into phase segregation and crack propagation dynamics [44]. This approach provides a nuanced understanding of mechanical stresses and electrochemical processes during battery operation, which are crucial for predicting LIB performance and lifespan.

Integrating real-time stress measurement techniques, such as those proposed by Sethuraman et al., enhances understanding of mechanical degradation mechanisms in composite electrodes [47]. These methods offer direct, quantitative insights into stress evolution within battery materials, informing the development of more robust designs. However, the complexity of battery degradation processes poses challenges for accurate Remaining Useful Life (RUL) prediction, as current studies struggle to obtain reliable data due to the multifaceted nature of these phenomena [6].

Non-invasive testing methods, including ultrasound and layer-resolved characterization, provide additional avenues for monitoring changes in states of charge and health during battery operation [2].

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These techniques enable non-destructive assessment of battery health, offering valuable data to refine models and improve degradation prediction accuracy. Despite advancements, the complex ultrasound response of second-life batteries underscores ongoing challenges in analyzing intrinsic defects and material stability [48].

Developing coupled phase-field formulations further addresses the limitations of traditional static models by providing a more accurate representation of crack behavior in LIBs [49]. This advancement facilitates better risk assessment and design strategies, contributing to safer and more reliable energy storage solutions. Models integrating ion and electron transport effects offer a comprehensive understanding of SEI growth dynamics and capacity fade, critical for optimizing LIB performance [30].

The intricate interactions among various degradation mechanisms in LIB systems highlight the need for ongoing enhancements in modeling techniques. These advancements are crucial for accurately representing complex, multifactorial degradation processes significantly impacting battery performance and lifespan. Recent studies have demonstrated limitations in existing models, which often fail to account for interplay among multiple degradation pathways within a single electrode. Research has successfully coupled four degradation mechanisms using the PyBaMM modeling framework, revealing diverse pathways to end-of-life scenarios based on usage patterns. Moreover, incorporating degradation mode analysis has emerged as vital for validating lifetime predictions, emphasizing the necessity of a comprehensive approach that encompasses various performance metrics beyond mere capacity and resistance. This multifaceted understanding is essential for optimizing battery design and enhancing reliability of energy storage solutions in pursuit of decarbonization goals [46, 18, 16]. By leveraging advanced analytical tools and innovative modeling frameworks, researchers can enhance predictive capabilities, ultimately supporting the development of more efficient and sustainable battery technologies.

5 Surface Reconstruction and Interface Modification

Exploring strategies for surface reconstruction and interface modification is essential to enhance the performance of lithium-ion batteries (LIBs). These strategies are crucial in optimizing the electrochemical properties and structural integrity of electrode materials. The following subsections discuss advanced approaches, such as phosphorus doping and graphene protrusions, which improve lithium-ion transport and contribute to the stability and longevity of LIBs.

5.1 Phosphorus Doping and Graphene Protrusions

Phosphorus doping and graphene protrusions represent sophisticated techniques for enhancing LIB electrode performance by improving electrochemical properties and reducing degradation. Phosphorus integration into graphene creates protrusions that enhance the interaction with lithium ions, facilitating efficient ion transport and charge storage, thus boosting electrode stability and battery longevity [5]. Graphene's conductivity and robustness make it an ideal substrate for phosphorus doping, optimizing the distribution of active phases and addressing voltage hysteresis issues [8].

Advanced imaging techniques, capable of extracting redox orbitals, provide insights into mechanical failure modes and stress management, enhancing understanding of surface interactions [33]. Recognizing defects like oxygen vacancies and manganese antisites is vital for tailoring surface properties to improve performance and stability [25]. Non-destructive visualization techniques offer real-time insights into battery deterioration, supporting surface reconstruction efforts [32].

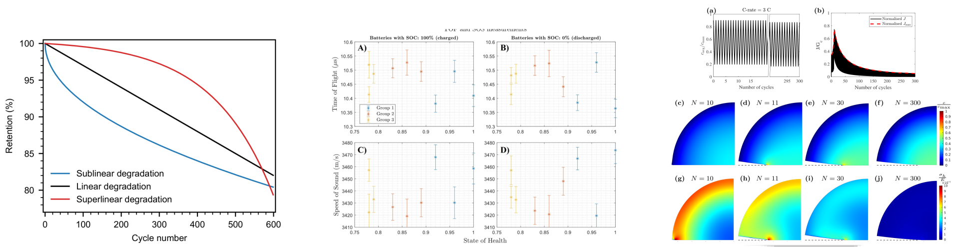
Overall, phosphorus doping and graphene protrusions significantly advance LIB design, enhancing lithium-ion interaction and preventing detrimental interfacial reactions, thus bolstering structural integrity and safety [5, 9, 27, 1].

5.2 Advanced Imaging and Modeling Techniques

Advanced imaging and modeling techniques, including X-ray computed tomography and operando optical tracking, are vital for understanding surface reconstruction processes in LIBs. These methods visualize phenomena like electrode fracture and ion dynamics, crucial for optimizing battery materials and designs to improve capacity retention and performance [34, 27, 50, 32]. Operando optical tracking provides accessible insights into ion dynamics, essential for studying phase transitions and optimizing active phase distribution [50].

Advanced modeling techniques, integrating electro-chemo-mechanical frameworks with phase-field models, predict damage in realistic battery structures, capturing interactions between reactions and stresses [34]. These models guide the design of resilient materials by simulating the effects of surface modifications on electrode stability.

Integration of imaging techniques with modeling frameworks allows comprehensive analysis of surface reconstruction, enhancing understanding of fracture mechanics and performance factors, paving the way for next-generation battery designs [34, 17, 50, 32].



(a) Degradation of Retention over Cycle Number[38]

(b) TOF and SOS measurements[48]

(c) Steady-state dynamics of a coupled oscillator system with a periodic external force[49]

Figure 5: Examples of Advanced Imaging and Modeling Techniques

As shown in Figure 5, surface reconstruction and interface modification are crucial in understanding complex systems. The examples illustrate studies leveraging advanced techniques for insights into battery aging, state of health, and oscillatory system behavior, underscoring the importance of imaging and modeling in unraveling material and system dynamics [38, 48, 49].

5.3 States-Filling Model (SFM) and Surface Reconstruction

The States-Filling Model (SFM) is crucial for understanding and enhancing electrochemical interactions at LIB electrode surfaces. It links lithium-carbon binding energy with work required to fill electronic states, optimizing carbon-based anode properties for performance and durability [29]. Integration with phase-field models simulates electrochemical behavior under stress, informing resilient material design [44].

High-throughput imaging, like operando optical tracking, provides insights into phase transitions and ion dynamics, aiding surface reconstruction strategies [50]. Coupling SFM with models like the Coupled Degradation Mechanism Model (CDMM) supports accurate degradation mode fitting across temperatures, crucial for understanding interactions in surface reconstruction and interface modification [18].

5.4 Atomic Layer Deposition (ALD) for Interface Modification

Atomic Layer Deposition (ALD) is a precise technique for modifying LIB interfaces, creating passivating layers to reduce decomposition and enhance stability [51]. Insights from density functional theory (DFT) inform surface property tailoring for improved ion transport and reduced degradation [9]. Integration with photonic crystal structures offers real-time insights into interface modification effects [52].

Advanced stress measurement methods provide data on mechanical behavior, optimizing ALD processes for durability [47]. Systematic feature design frameworks and deep transformer networks enhance predictions of battery health and cycle life, supporting efficient and sustainable technologies [27, 16].

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5.5 Cohesive Phase-Field Interface Model (CPF)

The Cohesive Phase-Field Interface Model (CPF) simulates fracture behavior in LIB materials, crucial for interface modification. Integrating cohesive zone models with phase-field methods, it captures fracture patterns and interactions, enhancing understanding of mechanical and electrochemical processes [45]. It maps relationships between degradation mechanisms, optimizing design and maintenance strategies [16, 43, 18, 2]. CPF supports resilient material design, enhancing LIB lifespan and performance.

5.6 Grain Boundary Engineering with Solid Electrolyte Infusion (GBE-IE)

Grain boundary engineering with solid electrolyte infusion (GBE-IE) enhances LIB cycle stability, especially in nickel-rich cathodes. Infusing lithium phosphate (Li_3PO_4) into grain boundaries mitigates side reactions, improving mechanical integrity and extending battery lifespan [27]. It prevents liquid electrolyte penetration, reducing capacity loss and voltage instability [27].

Solid electrolyte infusion facilitates uniform lithium-ion diffusion, crucial for consistent performance and preventing localized degradation. Enhanced ionic conductivity at grain boundaries improves cycling stability, addressing issues like capacity retention and voltage stability at various temperatures [27, 5, 7, 25, 53].

GBE-IE significantly advances LIB material design, enhancing cycle stability and voltage retention. By addressing grain boundary stability and electrolyte interactions, it supports durable and efficient energy storage solutions, aligning with sustainability and performance goals [1, 27].

6 Electrode Regeneration and Battery Sustainability

6.1 Concept and Importance of Electrode Regeneration

Electrode regeneration is a pivotal strategy in extending the lifecycle of lithium-ion batteries (LIBs), targeting the restoration of degraded electrodes to enhance battery longevity and sustainability. This method addresses electrode degradation, a key factor affecting battery performance and capacity, by revitalizing spent electrodes through advanced regeneration processes. This approach mitigates waste, recovers critical materials such as lithium, cobalt, and nickel, and aligns with global sustainability initiatives focused on resource conservation and minimizing ecological impact. The economic viability of these processes is further enhanced by innovative recycling technologies, ensuring a sustainable lifecycle for LIBs [13, 15, 19, 4, 12].

Various techniques are employed in the regeneration process to counter mechanical and chemical degradation effects. For instance, maintaining the structural integrity of SnO anodes is essential for sustainability, ensuring continued electrochemical performance [3]. Additionally, the infusion of Li_3PO_4 into the grain boundaries of Ni-rich NMC cathodes enhances structural and interfacial stability, leading to improved capacity retention and reduced voltage decay during cycling [27]. These strategies highlight the importance of structural optimization in electrode regeneration.

Optimizing lithium staging dynamics through mechanical strain is critical for battery sustainability [21]. Understanding defect interactions and site preferences in doped $LiMO_2$ materials enhances electrochemical performance [54]. Predictive maintenance strategies also underscore the importance of electrode regeneration for sustaining battery health and performance [6].

Through electrode regeneration, resource conservation is significantly enhanced, waste is reduced, and performance metrics are improved, addressing environmental concerns linked to LIB production and disposal. Recent advancements, such as the use of multifunctional organic lithium salts for direct regeneration of degraded cathodes, demonstrate the potential for restoring battery efficiency while minimizing ecological footprints. As LIB demand increases, particularly in electric vehicles, effective regeneration strategies will be essential for a sustainable energy storage future [13, 15, 19, 12, 43].

6.2 Techniques for Electrode Regeneration

Electrode regeneration techniques are crucial for extending the lifespan and enhancing the sustainability of lithium-ion batteries (LIBs). These methods focus on restoring the electrochemical

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performance of degraded electrodes, thereby reducing waste and conserving valuable materials. A primary approach involves rejuvenating electrode materials through structural optimization and defect management. For instance, infusing Li_3PO_4 into grain boundaries of Ni-rich NMC cathodes significantly enhances structural and interfacial stability, resulting in improved capacity retention and reduced voltage decay during cycling [27].

Another promising technique involves optimizing lithium staging dynamics through mechanical strain, leveraging the mechanical properties of electrode materials to enhance lithium-ion transport and reduce degradation. Understanding the impact of microstructural parameters on mechanical degradation enables the development of strategies to enhance electrode performance. Real-time stress measurement techniques, such as the wafer-curvature method, provide valuable insights into the mechanical behavior of electrodes during electrolyte wetting and electrochemical cycling [47]. Future research should integrate these stress measurement techniques with detailed 3D modeling to better understand microstructural interactions.

Advanced imaging and modeling techniques also play a crucial role in electrode regeneration. Methods such as operando optical tracking and electro-chemo-mechanical modeling offer insights into phase transitions and ion dynamics at the single-particle level, enabling researchers to visualize real-time changes in ion distribution and structural evolution, which inform effective regeneration strategies [50].

The advancement of electrode regeneration techniques is critical for improving LIB sustainability and performance. By emphasizing structural optimization through techniques like ultrasonic resonance for non-destructive characterization, coupled with advanced modeling that integrates multiple degradation mechanisms and effective defect management via innovative grain boundary engineering, these approaches significantly enhance the durability and environmental sustainability of lithium-ion batteries, paving the way for more reliable and eco-friendly energy storage solutions [2, 18, 27].

6.3 Integration of Regenerated Electrodes into New Battery Cells

Integrating regenerated electrodes into new lithium-ion battery (LIB) cells is pivotal for enhancing sustainability and efficiency in energy storage technologies. This process involves reassembling spent electrodes that have undergone regeneration to restore their electrochemical properties into new battery architectures. Effective integration of regenerated components is vital for extending operational lifespan, thereby mitigating environmental impacts associated with the extraction, production, and disposal of battery materials. This approach addresses the rising LIB demand driven by electric vehicles and offers a sustainable solution to waste management challenges, as recycled materials can serve as valuable secondary sources for essential elements like lithium, cobalt, and nickel used in battery manufacturing [15, 4, 18].

A key challenge in integrating regenerated electrodes is ensuring compatibility with existing materials and designs of new battery cells. This necessitates a thorough understanding of the mechanical and electrochemical properties of regenerated electrodes, achievable through advanced characterization techniques. For instance, operando optical tracking and electro-chemo-mechanical modeling provide insights into phase transitions and ion dynamics of regenerated electrodes, facilitating their seamless incorporation into new cells [50].

Moreover, structural optimization of regenerated electrodes, such as infusing Li_3PO_4 into grain boundaries, enhances mechanical integrity and interfacial stability, making them more suitable for integration into new battery cells [27]. This enhancement not only improves electrochemical performance but also ensures long-term stability within the new cell architecture.

The integration process also benefits from advanced stress measurement techniques, like the wafer-curvature method, which provides data on the mechanical behavior of electrodes during electrolyte wetting and electrochemical cycling [47]. Understanding mechanical interactions between regenerated electrodes and other cell components enables optimization of the assembly process to minimize stress-induced degradation and enhance overall battery performance.

Integrating regenerated electrodes into new LIB cells represents a pivotal advancement in battery technology, enhancing performance while providing a sustainable solution to resource conservation and waste reduction. This innovation addresses environmental challenges linked to extracting critical materials such as lithium, cobalt, and nickel essential for LIB production. Recycling methods,

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including hydrometallurgical and direct recycling processes, aim to minimize the ecological footprint of battery manufacturing and contribute to a circular economy, ultimately mitigating adverse impacts of battery disposal and resource depletion [15, 19, 12]. Leveraging advanced characterization, modeling, and structural optimization techniques supports the development of more durable and environmentally friendly energy storage solutions.

6.4 Challenges and Solutions in Electrode Regeneration

Electrode regeneration in lithium-ion batteries (LIBs) faces significant challenges, including material degradation and complex interactions among battery components, necessitating effective solutions to enhance sustainability and performance. As LIB demand rises, particularly with electric vehicle growth, addressing these challenges is critical to mitigate environmental impacts associated with mining and processing key materials like lithium, cobalt, and nickel while enhancing overall battery efficiency and lifespan [15, 1]. A primary challenge is mechanical and chemical degradation during battery cycling, which affects the structural integrity and electrochemical properties of electrodes, complicating the regeneration process as it requires restoring both mechanical robustness and electrochemical functionality.

Managing thermal stability and degradation mechanisms under severe operating conditions, such as overcharging, presents a significant challenge. Advanced analytical techniques, including operando mass spectrometry and synchrotron scattering, provide valuable insights into reaction mechanisms during overcharge conditions, enhancing understanding of thermal stability and degradation pathways [35]. Future research should apply these methods to investigate severe overcharging conditions and various battery chemistries, further elucidating their impact on electrode regeneration processes.

Another challenge involves optimizing microstructural parameters to prevent mechanical degradation and enhance lithium-ion transport. Techniques like infusing Li_3PO_4 into grain boundaries have shown promise in stabilizing electrode structures and improving interfacial stability, offering potential solutions to mitigate degradation [27]. Additionally, advanced stress measurement methods, such as the wafer-curvature technique, inform strategies to optimize structural integrity during regeneration [47].

Integrating advanced modeling frameworks, such as electro-chemo-mechanical models, enhances battery solution development by providing a detailed understanding of interactions between electrochemical reactions and mechanical stresses. These models elucidate how surface stresses in nano-sized anode particles influence mechanical endurance and electrochemical performance, revealing that while surface stresses may improve mechanical properties under specific conditions, they can simultaneously degrade electrochemical performance. Moreover, semi-empirical models tracking solid-electrolyte interphase (SEI) growth and cell thickness expansion during battery formation offer insights into optimizing manufacturing processes and predicting battery lifespan. Mechanical models accounting for gas-induced bulging in pouch-cell batteries allow accurate predictions of deformation and stress distribution, critical for assessing battery health over its lifecycle. Together, these comprehensive modeling approaches facilitate a deeper understanding of factors affecting battery performance and longevity [43, 40, 42]. They aid in predicting the impact of regeneration techniques on electrode performance and guide the design of more resilient materials.

Addressing challenges in electrode regeneration requires a multifaceted approach combining advanced analytical techniques, structural optimization strategies, and comprehensive modeling frameworks. By tackling significant challenges associated with LIBs, including environmental impacts of resource extraction and limitations of current recycling methods, researchers can develop more efficient regeneration processes. These advancements improve LIB sustainability by enhancing recovery of critical materials like cobalt, nickel, and lithium and optimize battery performance through innovations such as grain boundary engineering in cathode materials, contributing to a more circular economy in the growing electric vehicle market [15, 27, 12].

6.5 Impact on Battery Performance and Environmental Sustainability

Electrode regeneration significantly enhances the performance and environmental sustainability of lithium-ion batteries (LIBs) by restoring electrochemical properties of spent electrodes, thus extending battery lifespan and reducing waste. Techniques such as the Molten-Salt Direct-Recycling (MSDR) method exemplify sustainable recycling practices by regenerating high-performance nickel-rich cathodes from low-nickel sources, supporting closed-loop recycling initiatives for electric vehicle batteries [11]. This approach conserves resources while minimizing environmental impact linked to battery production and disposal.

Recent advancements in regeneration processes, including direct regeneration of $LiFePO_4$ cathodes, showcase potential for maintaining excellent cycling stability and rate performance, with significant capacity retention over extended cycles [13]. The integration of phosphorus doping in electrode materials enhances lithium adsorption, although challenges like lithium clustering must be addressed to optimize cycling performance and sustainability [9]. Understanding kinetic factors influencing voltage hysteresis is crucial for improving battery sustainability, as these factors significantly impact electrode design and performance [39].

Advanced characterization techniques, such as operando optical transmission spectra, provide valuable insights into electrochemical performance and structural integrity of LIB materials over extended cycling [50]. These insights are instrumental in optimizing regeneration processes and ensuring long-term stability of regenerated electrodes. Additionally, high-resolution x-ray Compton scattering offers direct insights into redox processes, crucial for understanding and improving battery performance [33].

Intrinsic defects significantly impact battery performance, influencing electrochemical properties of materials like $LiMnO_2$ and Li_2MnO_3. Nanostructuring and ion substitution strategies are recommended to enhance electronic conduction and overall battery performance [25]. The interaction of larger boron nitride flakes with lithium ions enhances stability and modifies electronic properties, positively impacting both battery performance and environmental sustainability [8].

Integrating predictive maintenance strategies that emphasize accurate health predictions is essential for sustaining battery performance and environmental sustainability [46]. By focusing on restoration and optimization of electrode materials, regeneration techniques not only enhance battery performance but also align with broader environmental goals by reducing waste and conserving resources. Continued research in this field will be crucial for developing effective regeneration strategies that support sustainable growth in energy storage technologies.

7 Case Studies and Recent Advances

7.1 Innovative Analytical Techniques in Recycling

Innovative analytical techniques have transformed lithium-ion battery (LIB) recycling, enhancing both efficiency and sustainability in material recovery. Operando mass spectrometry allows for real-time gas evolution analysis during battery operation, linking gas production to structural changes, thus optimizing recycling strategies by elucidating degradation mechanisms [35]. High-resolution x-ray Compton scattering (HR-XRCS) provides detailed insights into redox orbitals and electron momentum density, revealing subtle electron occupancy changes and guiding recycling methods that maintain the intrinsic properties of recovered materials [33]. Additionally, advanced modeling frameworks, such as the phase-field model, simulate interactions between electrochemical reactions and mechanical stresses, enhancing the design of resilient recycling strategies [44]. These techniques collectively advance LIB recycling by improving material recovery and reducing environmental impacts, supporting sustainable energy storage solutions amidst growing electric vehicle demands [27, 15, 19, 2, 12].

7.2 Advancements in Phase Transition Understanding

Recent insights into phase transitions in recycled lithium-ion battery (LIB) materials have significantly enhanced recycling efficiency and sustainability. These advancements are crucial as LIB demand surges, particularly for electric vehicles, necessitating innovative recycling technologies to mitigate environmental impacts [12, 15, 4, 19]. HR-XRCS has been pivotal in elucidating redox processes and electron momentum density, capturing subtle changes during battery cycling [33]. The phase-field model, which combines electrochemical and mechanical stress simulations, guides the design of resilient recycling strategies by accurately representing phase transition effects on material stability [44]. Operando optical tracking further enhances understanding of ion dynamics and phase transitions at the single-particle level, informing recycling process optimization [50]. These advancements not only improve recycling profitability but also contribute to a circular economy by providing secondary sources of essential materials for future battery production [4, 27, 12].

7.3 Successful Implementations in Direct Recycling

Direct recycling methods for lithium-ion batteries (LIBs) have demonstrated significant advancements in material recovery and sustainability. The Molten-Salt Direct-Recycling (MSDR) technique, for instance, successfully upcycles low-nickel polycrystalline cathodes into high-performance nickel-rich single-crystal cathodes, showcasing its economic and environmental viability [11]. Similarly, direct regeneration of spent $LiFePO_4$ cathodes using multifunctional organic lithium salts achieves excellent cycling stability and performance [13]. Techniques like infusing Li_3PO_4 into grain boundaries enhance structural stability, preventing degradation and improving the longevity of recycled materials [27]. HR-XRCS supports these implementations by providing insights into redox processes, ensuring material integrity during recovery [33]. These successful case studies highlight the potential of direct recycling to revolutionize LIB recycling practices, addressing environmental challenges and supporting a circular economy in battery resource management [15, 19, 12, 4].

7.4 Impact of Nanoscale Insights on Recycling

Nanoscale insights have significantly advanced recycling techniques for lithium-ion batteries (LIBs) by improving understanding of material properties and degradation mechanisms at the atomic level. As LIB demand increases, particularly in electric transportation, effective recycling methods are crucial to mitigate environmental pollution and resource waste [1, 27, 19]. HR-XRCS has played a pivotal role in revealing redox processes and electron momentum density, facilitating the development of targeted recycling methods [33]. Integrating nanoscale insights with advanced modeling frameworks, like the phase-field model, supports recycling optimization by simulating interactions between electrochemical reactions and mechanical stresses [44]. Techniques such as the Molten-Salt Direct-Recycling (MSDR) method utilize nanoscale properties to enhance material recovery and reduce environmental impact [11]. These advancements contribute to sustainable energy storage technologies by enhancing material recovery and reducing the environmental footprint of battery disposal, addressing challenges associated with LIB production and disposal [15, 2].

8 Challenges and Future Directions

The recycling of lithium-ion batteries (LIBs) faces complex challenges that require addressing technological constraints, economic feasibility, regulatory and safety issues, and the need for innovative material and design solutions. This section explores these multifaceted challenges and outlines future research opportunities to enhance recycling practices.

8.1 Technological Limitations

Direct LIB recycling is hindered by technological challenges, including the complexity of modeling solid electrolyte interphase (SEI) growth dynamics, which current models fail to accurately depict, leading to lithium consumption and capacity loss [30, 7]. The diverse chemistries of LIBs further complicate recycling, as many studies neglect the economic and technical complexities of different battery types, resulting in unreliable predictions [4, 18]. Achieving effective lithium adsorption on materials like boron nitride flakes without compromising safety remains challenging [8]. Moreover, the complexity of battery systems, particularly in electric vehicles, necessitates real-time monitoring to improve recycling outcomes [5]. Addressing these challenges requires sophisticated modeling, innovative materials, and real-time monitoring solutions to enhance the efficiency, safety, and sustainability of direct recycling methods [27, 15, 19, 4, 12].

8.2 Economic Feasibility

Economic challenges in direct LIB recycling include ineffective lithium recovery, essential for economic viability, and the complexity and cost of technologies needed for material recovery [12, 15]. Limited consumer awareness and inadequate recycling infrastructure further hinder collection and processing efficiency [15]. Integrating advanced modeling techniques, such as the cohesive phase-field interface model, complicates economic assessments [45]. To address these issues, developing cost-effective technologies and enhancing infrastructure for efficient LIB collection and processing is crucial. This approach will mitigate environmental impacts, recover valuable materials, and support the electrification of transportation as LIB demand rises [1, 15, 19, 4, 12].

8.3 Regulatory and Safety Challenges

Regulatory and safety challenges significantly impact LIB recycling. The lack of standardized guidelines leads to regional variability, complicating recycling practices and increasing operational costs [4, 15]. Safety risks arise from thermal runaway and hazardous material handling during disassembly and processing, necessitating stringent protocols and advanced technologies [8, 5]. Establishing comprehensive regulatory frameworks to standardize practices and ensure safety is essential. These frameworks should address environmental impacts, facilitate critical material recovery, and promote innovative recycling methods for sustainability and profitability [15, 4, 12].

8.4 Material and Design Innovations

Material and design innovations are crucial for improving LIB recycling outcomes. Optimizing synthesis conditions to control defect concentrations in materials like $LiMn_2O_4$ can enhance performance during recycling [55]. Chemical modifications to graphite surfaces and advanced electrolytes can reduce structural damage and improve stability [22]. Operando optical tracking techniques, such as iSCAT, provide insights into microstructural factors influencing recycling efficiency [50]. Infusing lithium phosphate into grain boundaries enhances structural stability in nickel-rich cathodes [27]. These innovations aim to improve recovery rates of valuable metals while addressing limitations of traditional methods, contributing to a sustainable LIB lifecycle [15, 19, 12].

8.5 Integration of Advanced Technologies

Advanced technologies present significant opportunities for enhancing LIB recycling efficiency and sustainability. Imaging and monitoring techniques, such as operando mass spectrometry and synchrotron scattering, provide real-time insights into reaction mechanisms, optimizing recycling strategies [35]. Advanced modeling frameworks, like the phase-field model, guide resilient recycling strategies by simulating dynamic interactions in LIB materials [44]. High-throughput screening technologies enhance safety and reliability, ensuring optimal conditions for material recovery [37]. Innovative material solutions, such as boron nitride flakes, improve stability and safety [8]. By leveraging these advancements, researchers optimize material recovery processes, addressing traditional limitations and maximizing key material recovery [27, 12].

8.6 Future Research Opportunities

Future research should focus on enhancing LIB recycling through several key areas. Exploring grain boundary strengths and electrolyte interactions can inform robust recycling strategies [45]. Improving recycling technologies, automating disassembly, and developing sustainable chemistries can mitigate environmental impacts [4]. Advanced imaging techniques offer potential for non-destructive visualization of internal battery conditions [32]. Integrating stress effects and electrolyte compositions into phase-field models will enhance predictive capabilities [28]. Studying SEI formation and optimizing model parameters will improve understanding of degradation and recycling mechanisms [26, 31, 18]. Focusing on these areas will drive progress in LIB recycling, enhancing material recovery efficiency and supporting the sustainable growth of energy storage technologies [15, 19, 12, 4].

9 Conclusion

The survey underscores the pivotal role of direct recycling methods in enhancing the sustainability of lithium-ion batteries (LIBs) by effectively addressing failure mechanisms and promoting efficient material recovery. Techniques such as Molten-Salt Direct-Recycling (MSDR) exhibit substantial promise in regenerating high-performance cathodes, thereby facilitating closed-loop recycling initiatives that mitigate environmental impacts [12]. These methods conserve valuable resources and significantly reduce the environmental footprint associated with battery production and disposal.

The necessity of developing efficient recycling processes is emphasized to improve recovery rates of lithium and other critical materials essential for the economic viability and sustainability of LIB technologies [12]. Furthermore, the survey highlights the importance of addressing safety issues through enhanced materials and design, as lithium-ion batteries are crucial for sustainable energy solutions [10].

Advanced analytical techniques and modeling frameworks have been instrumental in optimizing recycling processes, offering deeper insights into the structural and electrochemical properties of LIB materials. These innovations facilitate the development of more effective recycling strategies, ultimately contributing to the sustainable growth of energy storage technologies. By leveraging these advancements, direct recycling of LIBs can be executed more efficiently and sustainably, aligning with broader goals of environmental protection and resource conservation.

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