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Exploring the Impact of Prior Degradation on the Performance and Lifetime of Second-Life Li-Ion Batteries

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Second-life batteries play a crucial role in advancing the principles of the circular economy and promoting sustainability in the energy sector. This study explores the impact of prior degradation referring to the extent and nature of aging experienced during a battery's first-life application on the performance and lifetime of second-life Li-ion batteries (LIBs). Factors such as environmental sustainability and cost-effectiveness are key drivers for the utilization of second-life LIBs, with advancements in battery management systems, diagnostics, and refurbishment techniques enabling stakeholders to make informed decisions. This study explores various dimensions of first-life degradation, including cycling degradation, capacity fade, lifespan reduction, and electrolyte decomposition, emphasizing the importance of understanding these processes to assess their impact on second-life battery performance, optimize repurposing strategies, and ensure operational safety. Despite the progress made, there is still a need for further research to fully understand the cause and effect of the aging on the use case for second-life batteries. In the end, this understanding will maximize their potential for use in sustainable energy infrastructure.

1. Introduction

Second-life batteries represent a crucial component in the emerging landscape of sustainable energy-storage solutions. These batteries undergo a transition from their primary use in electric vehicles (EVs) or other applications to a secondary application, where they continue to contribute to energy-storage capacities. The second-life stage begins when a battery is deemed no longer suitable for its original use typically at ≈80% state of health (SoH) but still retains sufficient capacity for secondary applications.

The concept of second-life batteries aligns with the principles of the circular economy, aiming to extend the lifespan of resources and minimize waste generation by repurposing functional components. This approach not only addresses environmental concerns associated with battery disposal but also facilitates the development of a more sustainable and resilient energy infrastructure and decreases demand for resources, particularly rare-earth elements. First-life refers to the battery's initial deployment (e.g., in EVs), while second-life denotes the phase after retirement from that primary application. Degradation in the second-life stage builds upon, and is influenced by the degradation experienced in the first-life phase. "Second-life degradation" refers to the aging processes during this second-use phase, whereas "prior degradation" indicates the accumulated wear during the first life.[1]

By 2030, demand for lithium-ion batteries (LIBs) is projected to soar, requiring new value-chain strategies. Regulatory frameworks in the EU (e.g., EU Battery Directive) support recycling and reuse. The second-life market is driven by rising EV adoption, anticipated to generate over 3 million end-of-life battery packs by 2025. Since a Li-ion battery's usable life is limited, it is essential to comprehend the automotive battery life cycle. This problem is addressed by regulations for battery disposal and take-back, especially in the European Union, where strict legislation like the EU Battery Directive requires battery recycling and reuse.[2] In contrast, battery take-back initiatives in the US are state-level and lack comprehensive federal restrictions. Comparably, frameworks in Australia, Asia, and Africa are less standardized, with many areas managing battery end-of-life procedures through early-stage policies or voluntary initiatives. The process needed to recycle or repurpose second-life batteries depends on the specific battery type; LIBs require distinct procedures. LIB use, including second-life application, is a key component of policies to encourage the development of electric vehicles. Although there are currently considerable regional variations in regulatory methods, global standards are being developed to guarantee environmentally sustainable behavior within the battery business.[3]

The market for first life and second-life batteries is evolving rapidly, driven by the surge in EV sales, which are expected to reach 20 million units globally by 2025, contributing to a significant stockpile of end-of-life EV batteries, projected to exceed 3.04 million battery packs by then. Second-life batteries,

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repurposed for less demanding applications such as stationary energy storage and backup power systems, offer a cost-effective alternative to new batteries.[4]

Environmental sustainability stands as a prominent driver behind the growing interest in second-life batteries. By repurposing batteries that have reached the end of their automotive or renewable energy applications, significant reductions in carbon emissions and resource consumption can be achieved.[5] This approach mitigates the environmental impact associated with the extraction of raw materials, manufacturing processes, and disposal of batteries. Furthermore, it aligns with global efforts to transition toward a low-carbon economy by maximizing the utilization of existing resources and minimizing the reliance on finite natural resources.[6] **Figure 1** shows a typical life cycle of LIBs including a second-life phase.

Cost-effectiveness emerges as another compelling factor driving the adoption of second-life batteries. While these batteries may exhibit reduced capacity compared to new counterparts, they often present a more economical alternative for various energy-storage applications.[7] The lower upfront costs associated with repurposed batteries enhance the feasibility of energy-storage projects, particularly in scenarios where the primary concern is achieving a balance between performance and affordability. This cost-saving potential not only benefits individual consumers but also contributes to the broader accessibility and scalability of renewable energy technologies.[8]

Second-life LIBs face several challenges, including safety and reliability concerns due to wear and degradation, reduced capacity, and economic viability as the cost of new batteries decreases. However, advancements in battery management systems, their suitability for specific low-demand applications, and environmental benefits make these issues less critical. They extend battery life, reduce environmental impact, and are supported by ongoing improvements in recycling technologies.[9,10]

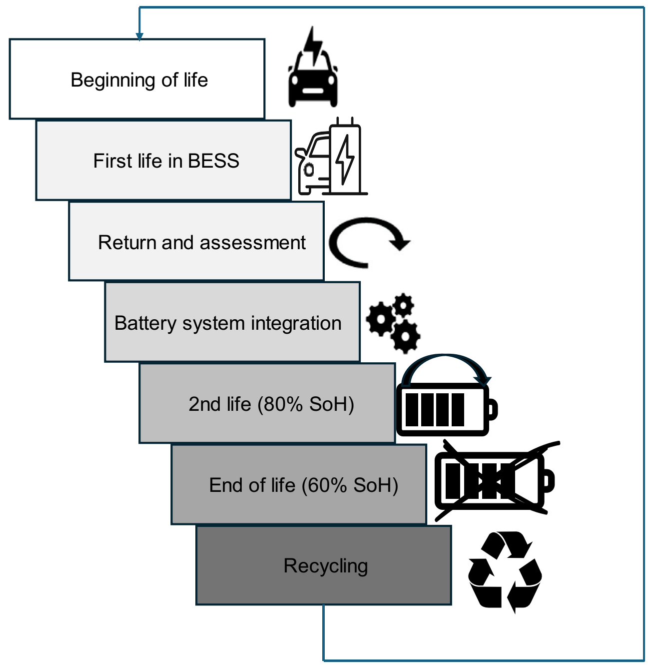

Figure 1. Lifecycle pathway of Li-ion batteries (LIBs) from initial deployment to recycling of LIBs. The second-life phase typically begins when the battery reaches the SoH of ≈80% and ends around 60% SoH.

Based on a dynamic degradation model of LIBs, Song et al. first compared the profits that second-life and fresh batteries could bring to wind farms. Two case studies in USA and Denmark were conducted and the analysis showed that given the current prices of wind energy and LIBs, reusing batteries was not cost-effective for the studied wind farms, but it may outperform fresh batteries in the future if the wind energy price decreases much faster than the battery price.[11] In another study, Mathews et al. found that when the cost of a second-life battery is

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less than 80% of a new battery, it becomes a more cost-effective option compared to using a new battery, especially when the project involves limiting the battery's state of charge (SoC) between 85% and 20%.[12]

Technological advancements play a pivotal role in unlocking the full potential of second-life batteries. With innovations in battery management systems, diagnostics, and refurbishment techniques, the process of assessing the health and performance of used batteries has become more precise and reliable.[13] These advancements enable stakeholders to make informed decisions regarding the suitability of repurposed batteries for specific applications, thereby maximizing their value and minimizing operational risks. Additionally, ongoing research and focus on enhancing the efficiency and longevity of second-life battery systems, further reinforcing their role as a viable and sustainable energy-storage solution.[14] LIBs pose elevated safety risks due to prior wear. Increased internal resistance in aged cells can cause localized heating and thermal runaway, leading to potential safety failures. Consequently, these batteries often fail to meet new safety certifications and require stringent management strategies such as temperature regulation and state-of-charge (SoC) limits. Tschirschwitz et al.[15] demonstrated that thermal runaway in second-life modules can result in severe consequences, including jet flames up to 5 m in length and significant mass loss of up to 82%. These findings underscore the critical need for comprehensive safety assessments and robust management protocols in second-life LIB applications.

The aging of batteries limits their total lifetime and the nonlinear aging behavior at later stages can diminish the benefit of second-life applications. A model-based aging study shows that the lifetime can be doubled by introducing a two-stage anode porosity.[16] Due to these factors, along with its inherent properties and widespread utilization in electric vehicles, it is anticipated that a considerable proportion of batteries taken out from EVs will be repurposed for secondary applications. In this scenario, CPFL Energy, in cooperation with CPQD and BYD Brazil, is developing an R&D project, to assess the remaining useful life of LIBs removed from EVs in a second application.[17] The assessment of the remaining useful life (RUL) of LIBs from electric vehicles is a critical aspect of battery management and prognostics. Rezvani et al. highlighted the importance of emerging prognostics and health management (PHM) techniques for accurately quantifying the SoH of LIB cells and predicting their remaining useful life.[18] This is crucial for ensuring the transparency of battery health assessment and predicting the impact on vehicle mobility. Sangwan et al. presented aging models and empirical capacity degradation modeling for re-used electric vehicle batteries, as well as the use of Kalman Filter to predict the remaining useful life of Li-ion batteries. These models and techniques are essential for accurately predicting the RUL of LIBs and ensuring their efficient use in second-life applications.[19] Depending on the precise circularity and impact groups taken into consideration, Life Cycle Assessment (LCA) studies have shown that recycled batteries generate 30–72% less environmental impacts and exhibit 13–47% increased circularity. Notably, these batteries can mitigate the effects of climate change by 16% and acidification by 25% when repurposed for stationary energy storage. This is mainly because they balance the efficiency loss compared to new batteries with the avoidance of new battery manufacture.[20] By analyzing multiple reuse scenarios, such as applications in energy-storage systems, communication base stations, and low-speed vehicles, in addition to various recycling techniques, such as hydrometallurgical, pyrometallurgical, and direct recycling, a recent study offers a way to optimize these pathways. The results show that, compared to direct hydrometallurgical recycling without prior reuse, incorporating reuse before recycling can increase revenues by 58% and reduce emissions by 18% for lithium-iron-phosphate (LFP) batteries. In a similar vein, lithium nickel manganese cobalt oxide (NMC) batteries demonstrate an 18% decrease in emissions and a 19% improvement in profit under the same circumstances. LFP batteries give better long-term benefits when reused before recycling, even if NMC batteries offer higher immediate recycling returns.[21]

Through the development of an equivalent circuit model, a numerical simulation was conducted by Philippot et al. by developing a system model that mimicked the behavior of the photovoltaic (PV) array and battery pack through the use of equivalent circuit methodologies.[21] The numerical investigation revealed that the proposed system, using second-life batteries, achieved similar performance to systems using new lithium batteries but at a reduced cost.[21] Another investigation[21] examined the viability of repurposing these batteries for second-life applications, aiming to facilitate the growth of intermittent renewable electricity sources in California until 2050. The study employs a comprehensive life-cycle system model that encompasses battery provisioning, degradation, logistical considerations, and utilization in second-life scenarios. The use of Ex transportation battery systems (i.e., second-life electric vehicle/hybrid electric vehicle batteries) in grid applications is an emerging field of study.[13] Another research compared all the modes of the converter along with their switching performances in detail to understand the relative advantages and disadvantages of each mode to help to select the suitable converter mode.[22] Other research focuses on the identification of the aging mechanism and estimation of the Soh of second-life batteries, using correlation-based feature selection methods, a universal index that is feasible for all batteries is presented for regression analysis, and the estimation error is found to be within 3%.[23] Casals et al. analyze the remaining useful life of second-life batteries on four different stationary applications, which are: Support to fast electric vehicle charges, self-consumption, area regulation, and transmission deferral. They study second-life batteries lifespan: rest of useful life and environmental analysis. This model runs in MATLAB and includes several aging mechanisms, such as calendar aging, C-rate, and depth of discharge (DoD). Proper disposal of spent batteries has always been a concern, but it has also been discovered that these batteries often retain enough energy perfectly suited for other uses, which can extend the batteries' operational lifetime.[24] Venkatapathy et al. aimed to identify the factors influencing the transition from the first life to the second-life of traction batteries. Their study highlighted cost, environmental considerations, and aging as key factors in this transition, with cost being the most significant. Their modeling framework, depicted in **Figure 2**, illustrates the interplay of these factors

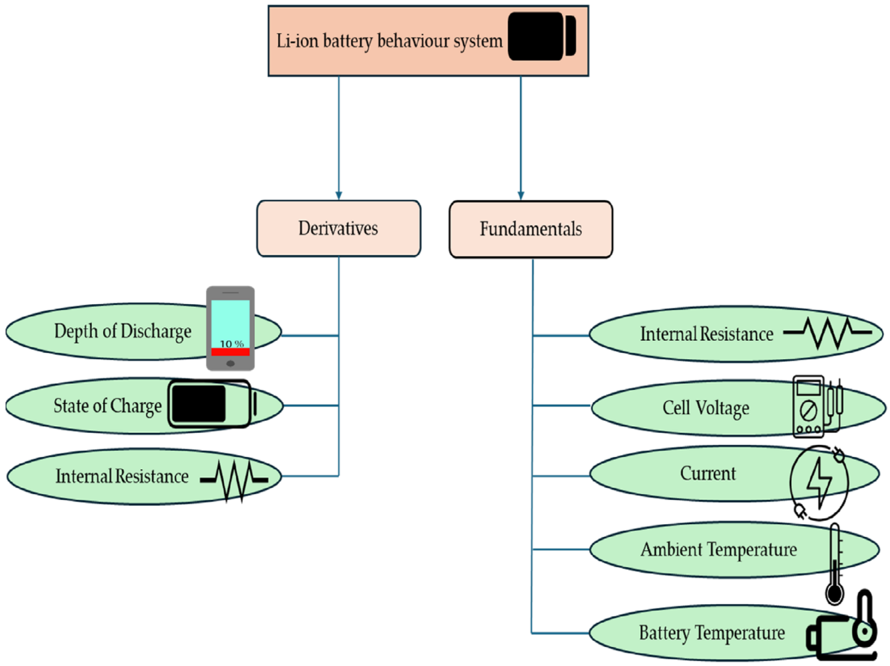

Figure 2. Framework to categorize LIB behavior by means of fundamental parameters (e.g., internal resistance, cell voltage) and derivative parameters (e.g., depth of discharge, SoC). This framework highlights the essential factors to determine a battery's usefulness for second-life application. Redrawn from ref. Reproduced with permission.[27] Copyright 2015, IEEE.

in determining the viability of second-life batteries compared to new ones.[25] In a study examining the potential of second-life hybrid vehicle batteries for solar backup systems, researchers investigated their suitability and durability for energy storage.[26]

Another study introduced the concept of reusing (EV) batteries to minimize carbon footprints and simplify recycling processes. Analysis of various stakeholders identified factors influencing the market diffusion of second-life EV batteries. The methods proposed for determining the optimal sizing of second-life battery energy-storage systems aim to enhance their economic viability.[27]

Overall, the benefits of second-life batteries are evident and have been demonstrated by numerous examples. Understanding the aging mechanisms of Li-ion batteries has advanced significantly, but not much is known about degradation processes particular to second-life batteries. The degree and rate of degradation in second-life applications of first-life batteries are significantly influenced by their aging and cycle history. Second-life performance is greatly influenced by variables like storage conditions, recycling procedures, and past usage history. Key aging processes such as electrolyte breakdown, capacity fading, and cycling degradations, are highlighted herein, along with their consequences for second-life operations. Our study offers an informed viewpoint on the difficulties and possibilities for repurposed batteries by describing the main degradation processes that occur during the first life and evaluating their effect on performance and longevity during the second-life. Additionally, discussions about battery management systems and diagnostic tools provide insightful information to enhance second-life applications, optimize battery design, and promote sustainable energy-storage options that are consistent with the circular economy.

The subsequent sections delve into aging mechanisms, such as chemical composition, providing in-depth analysis and insights into these critical aspects.

2. Aging Dimensions during Usage

The aging mechanisms of Li-ion batteries exhibit some variations between their first and second-life stages. In this study, we focus on the first use-case, which will contain degradation that the second-life will then inherit. In the initial use phase, common aging dimensions include capacity fade, internal resistance increase, electrode degradation, and side reactions.[28] These factors contribute to the gradual degradation of the battery's performance over time and charge cycles. On the contrary, in the second-life of LIBs, additional aging pathways such as previous use history, mismatched cells, stress from new applications, and storage conditions, can impact the battery's aging behavior.[29] While some aging pathways may persist or be similar between the two life stages, the specific factors influencing battery aging can significantly differ based on the application, usage patterns, and environmental conditions behavior.[29] Understanding these aging mechanisms is crucial for predicting the performance and longevity of LIBs in various applications. However, their performance and longevity are not immune to degradation over time and are attributed to a range of complex aging mechanisms. The formation of a degraded solid electrolyte interface (SEI) in the formation step, loss of active material, and structural breakdown caused by cycling are some of the causes underlying the degradation of LIB electrodes. These processes result in a decrease in capacity and cycling stability, as well as an increase in internal

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resistance. Electrode wear is also a result of chemical processes, including oxidation and lithium dissolution. Developing more effective and long-lasting energy-storage devices requires an understanding of these principles in order to improve battery design and longevity.[30]

Understanding these mechanisms is essential for optimizing battery design, enhancing performance, and ensuring safety.[31–33] To gain a comprehensive understanding of the complex aging dimensions in LIBs, we consider a range of research articles that clarify various degradation processes. These include cycling degradation, SEI formation, capacity fade, calendar aging, electrolyte decomposition, and particle cracking, each of which plays a critical role in the overall aging behavior of the batteries. It is important to emphasize that the initial use case will exhibit degradation, which the second-life will subsequently inherit.

2.1. Cycling Degradation

Repeated charge and discharge cycles induce both physical and chemical transformations within the battery's electrodes and electrolyte, leading to the degradation of electrode materials during lithium insertion and extraction. This degradation, common to many LIB chemistries, manifests as loss of active material, changes in electrode morphology, and increased resistance at the electrode–electrolyte interface.[33] Among various battery chemistries, $LiFePO_4$-based batteries have been widely studied due to their promising performance and unique degradation patterns. In this context, Cao et al. specifically explored the cycle-life and degradation mechanisms of $LiFePO_4$ batteries, identifying the loss of active lithium as a primary driver of aging.[34]

2.2. SEI Formation

During cycling, the SEI forms on the electrode surfaces due to electrolyte decomposition. While the SEI layer is essential for battery stability, its continuous growth and evolution can lead to capacity loss, increased impedance, and reduced cyclability.[35] Zhang et al. identified the formation reaction of the SEI film as a key cause of degradation, particularly at high ambient temperatures.[36] Xu et al. further explored the degradation of the crystal structure in lithium nickel oxide cathodes, which significantly reduces the redox activity of nickel and contributes to cycling performance decay.[37]

2.3. Capacity Fade

Capacity fade refers to the gradual loss of a battery's charge storage capacity over time. It occurs due to irreversible chemical reactions, structural degradation of electrode materials, and the accumulation of inactive components within the cell, such as the SEI layer.[38] As capacity fades, the internal resistance of the battery typically increases. Higher internal resistance leads to voltage drops during discharge, reduced power output, and diminished efficiency, further degrading battery performance.[39] Capacity fade can exacerbate safety risks associated with LIBs.[40] As the battery's capacity diminishes, it may become more prone to overcharging, over-discharging, and thermal runaway events, increasing the likelihood of safety hazards such as overheating, fire, and explosion.[14] Capacity fade results in a decrease in the battery's energy-storage capacity over time. As a result, the battery can store and deliver less energy per charge cycle, leading to diminished device runtime and overall performance.[41]

2.4. Calendric Aging

Even when not in use, LIBs experience chemical reactions that lead to capacity loss and performance degradation. This process, referred to as calendar aging, is driven by various factors, including temperature, humidity, electrode composition, and electrolyte formulation.[42] As shown in **Figure 3**, the cycling behavior of 18 650 cells is shown to highlight the impact of elevated temperatures on the degradation process of LIBs. Elevated temperatures significantly accelerate the aging process of 18 650 cells by facilitating side reactions and decomposition processes. This behavior is vital to better understand the degradation processes, as increased thermal stress contributes to faster degradation of cell performance and durability, which is crucial for optimizing battery management and improving reliability.[43] In[42] experimental observations, degradation mechanisms and modeling efforts have been discussed to predict the calendar lifetime of LIBs accurately and optimize their performance. The article emphasizes the importance of understanding degradation pathways and mechanisms to improve battery lifetime, especially in electric vehicles where battery management is crucial for maximizing their lifespan and efficiency.

2.5. Electrolyte Decomposition

High voltages and elevated temperatures are critical factors in accelerating the decomposition of electrolytes in LIBs. This process leads to the formation of gas bubbles, heat generation, and the depletion of electrolyte components, which in turn contribute

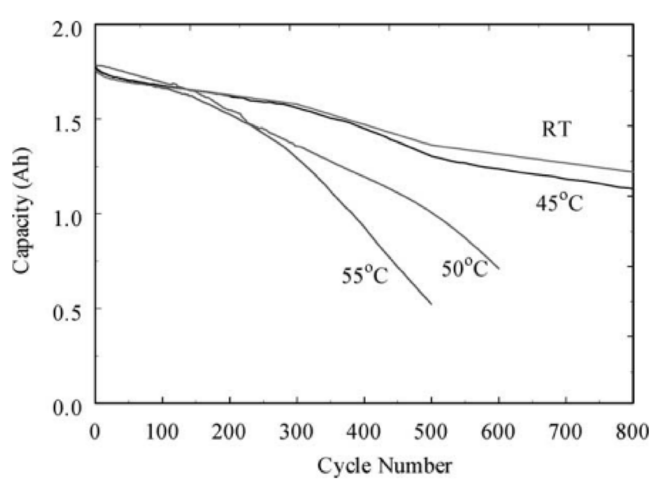

Figure 3. Discharge capacity of Sony 18 650 cells cycled at different temperatures as a function of cycle number. The graphic is reproduced with permission.[43] Copyright 2002, Electrochemical Society.

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to capacity fade, growth of the SEI, and increased safety risks like thermal runaway. At temperatures exceeding 80 °C, the SEI layer becomes unstable, initiating exothermic reactions between the lithiated carbon and the electrolyte. As temperatures rise beyond 100 °C, this degradation intensifies, leading to the release of gases and the melting of separators, with catastrophic consequences occurring around 135 °C.[44]

2.6. Particle Cracking

Mechanical stresses during battery cycling, particularly during the lithiation and delithiation processes, lead to microstructural damage in electrode particles. This damage manifests as cracking, fracturing, and loss of electrical contact, resulting in increased resistance, capacity loss, and overall mechanical degradation. As particles crack, new surfaces are exposed to the electrolyte, promoting further SEI formation and accelerating degradation. The combination of mechanical wear and chemical reactions at these newly exposed surfaces contributes significantly to the aging process of LIBs.[45]

2.7. Summary of Aging Dimensions

The aforementioned aging mechanisms affect the battery's lifetime, ultimately determining its performance, capacity retention, and cycle life. To address these degradation issues, a multifaceted approach is required, encompassing materials optimization, improvements in cell design, control of operating conditions, and advanced battery management systems.

Han et al.[46] specifically investigated the cycle life of commercial LIBs in electric vehicles to estimate capacity loss under real-world conditions. Their study involved designing battery cycle life experiments that mirror actual EV operating environments and using genetic algorithms to construct and fit battery cycle life models based on the observed aging mechanisms. As the demand for LIBs grows, especially for industrial and secondary applications, it becomes increasingly important to consider aging effects. This comprehensive understanding is crucial for addressing the safety concerns associated with extending the lifespan of these batteries. Based on the safety and dependability of using LIBs, four types of essential aging impact(s) were systematically identified.[47] These categories include degradation of construction integrity, self-heating effect, decay of thermal stability, and failure in key components. Life cycle experiments were carried out to determine how a large format $LiFePO_4$ battery's aging mechanisms are affected by the charging current rate and the cut-off voltage limit at a low temperature (−10 °C).[48] The aging mechanism is quantified using a mechanistic model, whose parameters are estimated with the particle swarm optimization algorithm (PSO). Wang et al.[49] study capacity-loss diagnostics and lifetime prediction in LIBs. They developed a capacity-loss diagnostic method based on open-circuit voltage analysis. A multi-mechanistic, non-destructive diagnostic technique is devised, using the open-circuit voltage (OCV) of a cell as the diagnostic sign. LIB aging mechanisms and life models under various charging stressors are studied by Gao et al.[50] Life cycle tests are used to determine how various cut-off voltages and charging current rates affect the battery aging process. It is also a crucial challenge for energy-storage systems to predict RUL and diagnose the SoH of batteries due to the complicated aging mechanism. For instance, Wei et al.[51] studied remaining useful life prediction and SOH diagnosis for LIBs using a particle filter and support vector regression. A novel method for battery RUL prediction and SoH estimation is proposed. Jiang et al.[25] focus on the identification of the aging mechanism and estimation of the SoH of second-life batteries. A universal index that works for all batteries is provided for regression analysis using correlation-based feature selection techniques, and the estimation error is discovered to be within 3%. To minimize the overall cost of energy consumption and battery degradation, Zhang et al.[52] developed an online, coordinated optimization strategy for a single-shaft parallel plug-in hybrid electric bus based on Pontryagin's minimal principle. First, a capacity loss model for LIBs emulating dynamics of both cycle life and calendar life is exploited in the optimization framework in order to highlight the importance of considering calendar life and its implication to overall energy management performance in real bus operations. From the standpoint of data-driven, battery curve matching, and recession characteristics for various applications, Ma et al.[53] suggest three distinct approaches for the capacity estimation problem of cells in series-retired battery modules. They contrast the applicability and limitations of every method based on the retired battery test data after deep cycling aging. Li et al.[54] use the incremental capacity (IC) curve to estimate the residual capacity of waste power batteries. Finally, through the analysis of the IC curve, a method for identifying the aging mechanism of large-scale decommissioned batteries is obtained.

Table 1. Classification of prior degradation severity.

Severity	Typical indicators	2nd-life suitability
Low	SoH > 85%, stable IR	High
Moderate	75% < SoH < 85%, mild Li plating	Medium
High	SoH < 75%, signs of severe Li plating or cathode cracking	Low to unsafe

As a summary **Table 1** categorizes LIBs based on their degradation severity, using typical indicators such as SoH, internal resistance, and signs of lithium plating or structural damage. This classification helps assess the suitability of cells for second-life applications, where batteries with low degradation are considered highly suitable, while those with severe degradation may pose safety concerns.

3. Factors Influencing the Aging of LIBs

The aging of LIBs can be influenced by various parameters, including: 1) Temperature: Elevated temperatures accelerate degradation processes within the battery, leading to faster aging. High temperatures can promote side reactions, increase electrolyte decomposition, and cause mechanical stress on electrode materials; 2) Mechanical stress: Physical factors such as mechanical strain,

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vibration, and electrode expansion/shrinkage during cycling can contribute to electrode degradation and loss of performance; 3) Chemical composition: The choice of electrode materials, electrolyte additives, and separator properties can influence battery aging rates. Optimizing material compositions and battery design can mitigate degradation mechanisms. For example, storage conditions. Prolonged storage at high temperatures or high states of charge can accelerate aging even when the battery is not in use. Proper storage conditions, including temperature and SoC management, are essential to minimize degradation during storage periods; 4) Cycling conditions: The number of charge–discharge cycles, as well as the DoD and charging rates, can impact battery aging. Cycling at high rates, deep discharges, and frequent cycling can accelerate degradation; 5) Voltage limits: Operating the battery beyond specified voltage limits, such as overcharging or over-discharging, can induce degradation mechanisms, including electrolyte decomposition, electrode swelling, and capacity loss; 6) State of charge: Operating the battery at high or low states of charge for extended periods can accelerate aging due to increased stress on the electrodes and electrolytes.[55–57]

3.1. Temperature

A method for calendar aging quantification of power batteries, taking into account the SoC and temperature effects, is presented in ref. [58]. The authors focused on a single parameter identified from electrochemical impedance spectroscopy (EIS) tests performed according to a specific protocol regarding thermal and electrical kinetics. For a LIB to function well and have a long service life, there must be little sudden change over time, in temperature as well as an appropriate operating temperature range. The heat generation and dissipation of LIBs are firstly analyzed based on the energy conservation equations, followed by an examination of the hazardous effects of an above-normal operating temperature. Variable parameters like electrode thickness and particle size of active material, along with optimization methods such as coating, doping, and adding conductive media are discussed in the electrode modification section, while the current development in air cooling, liquid cooling, heat pipe cooling, and phase change material cooling systems are reviewed in the thermal management part. In terms of heat sources, most of the heat produced by batteries originates from their electrodes; therefore, in order to effectively address the thermal problem with LIBs, the electrodes must be modified to achieve high overall ionic and electrical conductivity. To optimize the thermal properties of the complete cell, electrode modification can be achieved by reducing the thickness of the electrode and the size of the active material particles. This change in size will shorten the distance that ions travel through the electrodes and the active particles, respectively. Nonetheless, a battery with too thin electrodes will have a low active material loading.[59] In another study, authors used an electrochemistry-based model (ECBE) to investigate the aging behavior of LIBs operated within the temperature range of 25–55 °C. The results showed that higher temperatures accelerate the degradation of LIBs, primarily due to degradation mechanisms at the electrodes. In particular, the formation and modification of surface films on the electrodes, as well as structural/phase changes of the cathode, were found to be the main contributors to the increasing degradation rate of LIBs with temperature.[60] Another study[61] investigated the impact of high-temperature aging on the thermal safety of LIBs. The results showed that high-temperature aging can lead to increased heat generation and reduced thermal stability, which can increase the risk of thermal runaway. The study also found that the type of electrolyte used in the LIB can affect its thermal stability after high-temperature aging.[61]

Fleckenstein et al.[62] investigate how temperature gradients affect the aging of LIBs. They found that these gradients can accelerate degradation due to the uneven distribution of heat and current density within the battery cell. This inhomogeneous distribution exacerbates wear and tear, speeding up the battery's aging process. The study also emphasized that effective thermal management strategies can help mitigate these negative effects by improving the uniformity of heat and current distribution, thereby reducing the overall impact of temperature gradients on battery aging.

Deng et al.[63] summarize the latest research efforts on battery liquid cooling systems from three aspects, including the performance of coolant, classification of the liquid cooling system, and design of the battery pack. A major finding is that optimizing cooling performance and system design can significantly enhance the effectiveness of thermal management, thereby improving battery longevity and efficiency. Martinez-Laserna et al.[64] aim to evaluate the effects of Li-ion nickel manganese cobalt/carbon (NMC/C) battery SoH and aging history over the second-life performance on two distinct applications: one for power smoothing and renewable integration, and the other for home demand control. The thermal runaway performance of LIBs not only depends on materials and cell design but also changes with degradation. Ren et al.[65] present a comparative investigation of the aging effects on the thermal runaway behavior of a large-format LIB. Kim et al.[66] review the heat generation phenomena and critical thermal issues of LIBs. A novel battery thermal management system is proposed to provide an effective thermal management solution for high-energy density LIBs. Ouyang et al.[67] investigate the safety and chemical risks related to the use and mishandling of LIBs in electromobility. as is shown in **Figure 4** the design of the degradation test. All

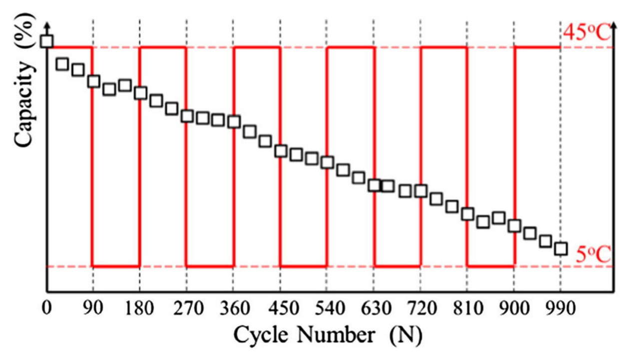

Figure 4. The design of the degradation test. The graphic is reproduced with permission.[67] Copyright 2016, Applied energy.

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batteries were fully charged at 1/3 C, and fully discharged at 1.5 C within one cycle using a Digatron tester inside a thermal chamber. The environmental temperature inside the thermal chamber switches from 45 C (5 C) to 5 C (45 C) after every 90 cycles. They identify and analyze the primary chemical and physical changes that occur when LIBs are exposed to thermal stress, whether from accidents or deliberate damage. Higher temperatures accelerate degradation mainly through mechanisms at the electrodes, including surface film formation and structural changes. Additionally, temperature gradients within cells cause inhomogeneous heat and current distribution, further hastening degradation. Effective thermal management strategies, like electrode modification and advanced cooling systems, are essential to mitigate these issues. However, there is a lack of comprehensive studies on the long-term impacts of these thermal management techniques and the combined effects of various aging conditions on battery safety and performance. Figure 4 illustrates the evolution of battery capacity over cycling under alternating temperature conditions. The black curve shows a steady decline in capacity with increasing cycle number, while the red step function represents periodic thermal cycling between 5 and 45 °C.

3.2. Mechanical Stress

Millner et al.[68] focus on modeling LIB degradation in EVs. A new aging model for LIBs is proposed based on theoretical models of crack propagation. From arbitrary charge and discharge histories, a stress metric for mixed use in cars or vehicle-to-grid activities is obtained. Cannarella et al.[69] propose a novel method of SoH/SoC determination using mechanical measurements. They present the results of long-term aging studies in which they observe stack stress to be linearly related to cell SoH for cells aged with different cycling parameters. The SEI fracture model of mechanical degradation in LIBs during cycling is studied by the authors in ref. [70]. A SEI layer encircling a spherical graphite particle serves as the basis for a mechanical model that is suggested. The model results were compared against cycle life aging experimental data, reproducing accurately the influence of the depth of discharge as well as the average SoC on the capacity fade. Xu et al.[71] propose a semi-empirical LIB degradation model that assesses battery cell life loss from operating profiles. For example,[55] developed a model by integrating basic ideas of battery degradation with the findings of battery aging tests. Sarkar et al.[72] study the mechanical and thermal aspects of lithium battery electrodes. Mechanical deformation was computed by combining the stress equilibrium equations with the electrochemical diffusion of Li-ions into the electrode particle. Second-life LIBs from EVs make up this prototype. The three applications of peak shaving (PS), German Primary Control Reserve (PCR), and uninterrupted power supply (UPS) are selected based on different stress scenarios of the battery energy-storage system (BESS), including operation mode as well as operation time. Hong et al.[73] explore how different parts of large-scale flexible LIBs age unevenly, using both micro-structural analysis and simulations. Their study reveals that in the battery design they analyzed the usage rates of electrodes and temperature distribution vary significantly. Specifically, they found that mechanical stress near the battery's connection tab is 8% higher than at the bottom edge. This increased stress near the tab is believed to accelerate aging in that area by about 35% compared to other parts of the battery. In [74] there is a focus on a physics-based aging model for LIBs with coupled chemical/mechanical degradation mechanisms. Battery degradation dynamics are described by an aging model based on physics. Then, the crack propagation due to the stress generated by the volume expansion of the electrode particles during cycling is modeled using Paris's Law of mechanical fatigue.[75]

Research on mechanical stress and aging in LIBs highlights several key findings. For instance, stress metrics from charge/discharge histories can be used to model degradation patterns over time. Mechanical measurements can determine SoC. Furthermore, fractures in the SEI layer contribute significantly to capacity fade. However, the impact of mechanical stress on long-term battery performance, especially under varied operational conditions, requires further exploration. Additionally, the interactions between mechanical and other degradation mechanisms, like thermal and chemical factors, remain under-researched, necessitating comprehensive models for accurate lifespan predictions.

3.3. Chemical Composition

In recent years, comprehensive battery research has been conducted, mainly aiming at better understanding the primary degradation processes occurring in these layered transition metal oxides. Both Ni-rich and Li-rich layered oxides of the NCM type are gaining a lot of attention as high-energy-density cathode materials for LIBs in order to meet the energy demands of the electromobility market. During formation and continuous cycling, especially, the electronic and crystal structure suffers from various changes, eventually leading to fatigue and mechanical degradation.[74]

Zhuang et al.[76] use attenuated total reflection geometry and Fourier transform infrared spectroscopy to analyze the surfaces of graphite anodes removed from Li-ion cells. The intricate chemistry of passive film production in actual Li-ion cells is reflected in the composition's multiple components. The recent interest in full and hybrid electric vehicles powered with Li-ion batteries has prompted in-depth battery investigations. By investigating the surface changes and passive film formation on anodes, researchers can gain insights into how these factors contribute to battery aging and overall performance, which is crucial for improving battery durability and efficiency in these applications. Particularly, the electrical and crystal structure experience a variety of changes during production and continual cycling, which ultimately results in mechanical and fatigue degradation.

Recent studies on different chemistries highlight significant changes in electronic and crystal structures during formation and cycling, leading to mechanical degradation and fatigue.[77] Techniques like Fourier transform infrared spectroscopy reveal the complex chemistry of passive films on graphite anodes, emphasizing the need to address these degradation issues to enhance the durability of batteries for electric vehicles. NMC,

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LFP, and NCA chemistries each exhibit unique degradation pathways. NMC degrades due to transition metal dissolution[78]; LFP is more stable but prone to mechanical cracking. NCA suffers from microstructural instability. Recent studies have confirmed these chemistry-specific degradation behaviors in detail. Wittman et al.[79] investigated the temperature-dependent degradation of commercial NMC and NCA lithium-ion cells and found that both chemistries exhibited increasing SEI growth and gas generation at elevated temperatures, while lithium plating was more dominant under low-temperature conditions. **Figure 5** shows how different chemistries show different aging behavior. To reach the 80% capacity cut-off, it took ≈170 EFC at 15 °C, ≈460 EFC at 25 °C, and over 600 EFC at 35 °C. By contrast, the NCA cells demonstrated no significant dependence on temperature. Ali et al.[80] examined the calendar aging of multiple battery chemistries, including LFP, NMC, and NCA, and demonstrated that LFP cells, while more thermally stable, showed gradual capacity loss due to structural strain and contact loss during long-term storage, in contrast to the more complex side reactions observed in NMC and NCA.

3.4. Cycling Condition

Petit et al.[81] study the development of an empirical aging model for LIBs and applications to assess the impact of vehicle-to-grid strategies on battery lifetime. An empirical capacity fade model for LIBs has been developed, calibrated, and validated for an NCA/C and an LFP/C Li-ion cell. The stress factors taken into account for each aging mode are the SoC and the temperature for calendar aging and the temperature and the current for cycle aging. The aim of these tests (electric, thermal, and aging) is to make a comparison between Li-ion technologies and choose the best one for each application.

Moreover, in ref. [82] 24 high capacity (1360 mAh) NMC622/Si-alloy Li-ion full pouch cells with high silicon-alloy content (55%) are cycle-aged under seven different cycling conditions to study the effect of different stressors on the cycle life of Si-anode full cells, among which are the effect of ambient temperature, DoD and the discharge current. A key result from this study is that optimizing the battery size, DoD, and energy management can significantly enhance the cycle life and performance of these cells. To maximize the cells' cycle life, energy content, and power output, the cells are also subjected to volumetric constraints at an ideal starting pressure, which helps in balancing durability and performance. For plug-in hybrid electric vehicles, Xie et al.[83] examine aging-aware co-optimization of battery size, DOD, and energy management. Convex programming is used to co-optimize battery size and energy management for plug-in hybrid electric vehicles (PHEVs), taking battery degradation into account. In this article, sensitivity analysis is also conducted to explore the effect of battery price and initial SoC on the optimal DoD and total cost. As shown in Figure 5, as the DoD increases, battery reaches faster to the end of its first life (80% SoH).

Pop et al.[84] studied the adaptive SoC indication system for LIB-powered devices. The overpotential model includes a variety of parameters that change during the cycling of the battery. Adaptive methods for the battery maximum capacity and for the overpotential model parameters used in the system with the aging effects are presented in this article. A range of lithium salts is present in the SEI, which is created during the initial cycles of life in LIBs and has a direct impact on the battery's aging performance. The behavior of high-power Li-ion cells over nearly three years is studied by Rizoug et al. through a combination of electric, thermal, and aging modeling, using an actual driving cycle. The study involves different configurations of tin (Sn) and carbon (C) films deposited as planar anodes for LIBs. The findings indicate that the choice of an anode material significantly impacts the performance and aging of the cells. Specifically, the study reveals that Sn-based anodes show a higher capacity retention and lower degradation rates compared to traditional carbon anodes under real driving conditions. Additionally, the research demonstrates that accurate modeling of electric, thermal, and aging effects can help in predicting battery performance more reliably and optimizing the design of high-power cells for electric vehicles. This comprehensive approach provides valuable insights into how different anode materials and operating conditions affect the long-term performance and durability of LIBs.[85]

Cycling conditions, such as ambient temperature, DoD, and discharge current, significantly impact the aging of LIBs. Higher temperatures and deeper discharge cycles accelerate degradation due to increased stress on battery materials. Research

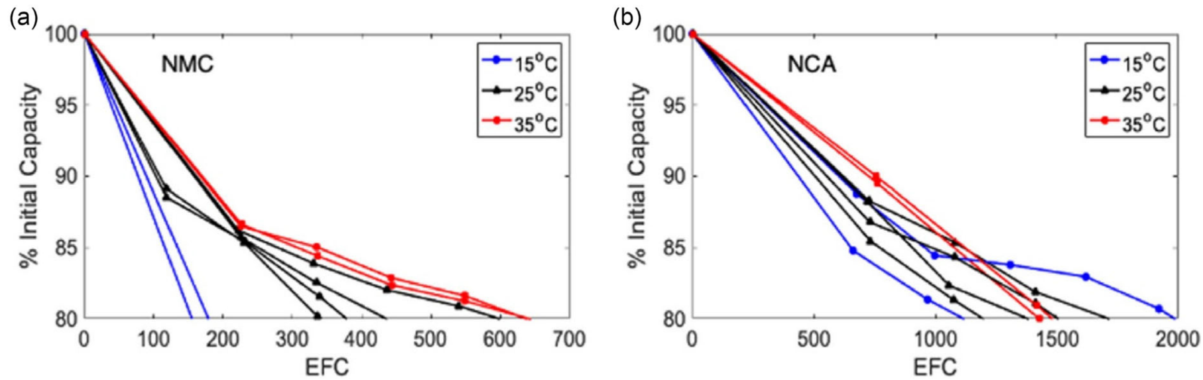

Figure 5. Discharge capacity fade based on equivalent full cycles (EFC) as a function of temperature for a) NMC and b) NCA cells in the original cycling study. The graphic is reproduced with permission.[79] Copyright 2023, Electrochemical Society.

shows that optimized cycling conditions can extend battery life by reducing mechanical and chemical wear. For instance, controlled temperatures and moderate DoD levels help maintain structural integrity and minimize capacity fade. Understanding these factors is crucial for enhancing battery longevity and performance in various applications, including electric vehicles and grid storage systems. **Figure 6** shows the effect of DoD on battery degradation, expressed as SoH over full equivalent cycles. Cells cycled at 100% DoD (blue line) exhibit the fastest degradation, dropping below 80% SoH in fewer than 160 cycles. In contrast, cells cycled at 60% DoD (yellow line) maintain over 80% SoH beyond 225 cycles, indicating significantly improved longevity. The trend demonstrates that reducing DoD during cycling effectively mitigates degradation, thereby extending battery life and enhancing suitability for second-life applications.

3.5. Impact of Previous Life Battery Degradation

Most researchers agree that prior degradation is the primary determinant of second-life viability, but debate exists around how best to measure or model it.[86] SoH alone is widely considered inadequate. Instead, a combination of electrochemical signatures (e.g., impedance, voltage curve shape) and advanced diagnostics (e.g., EIS, XRD) is recommended. Diagnostic methods are evolving, with Kalman Filters, OCV analysis, and data-driven machine learning models gaining traction for estimating RUL. The prior degradation conditions of LIBs are influenced by a multitude of factors including temperature fluctuations, DoD, charge/discharge rates, and usage patterns during the first life. For example, high-rate charging and deep discharges accelerate mechanical stresses and promote side reactions such as SEI layer growth, which contribute to capacity loss and impedance rise. Moreover, temperature extremes during first life can cause uneven degradation across cells within the same pack, leading to heterogeneous aging and imbalanced performance in second-life applications. These complex and interacting factors make it challenging to predict second-life behavior based solely on residual capacity or SoH metrics, emphasizing the need for detailed diagnostics and history-aware battery management systems to optimize reuse strategies.[87]

The primary causes of degradation in the first life of EV batteries have a major impact on the degradation behavior in their second-life. Cells with similar levels of SoH in their first life, especially those between 80% and 100% SoH, show similar rates of degradation when cycled under realistic second-life duty cycles. Even under the same cycling settings, the cells showed varying rates of degradation in their second-life when the underlying causes of degradation in the first life differed. This trend emphasizes how crucial it is to comprehend the precise causes of degradation in the first life since they have a direct bearing on the battery's performance and longevity in the second-life. Postmortem analysis further approved that cells with similar first-life degradation cause similar physical conditions and degradation patterns in their second-life, highlighting that SoH analysis alone is insufficient for predicting second-life performance In addition, degradation modes such as lithium plating, active material loss, and electrolyte decomposition do not progress uniformly but depend heavily on specific usage histories. Cells exposed to repeated fast-charging cycles are more prone to lithium plating, which can significantly compromise safety and reduce cycle life in their second life. Meanwhile, batteries operated predominantly at moderate temperatures and under shallow cycling tend to retain better structural integrity and electrochemical stability. Hence, understanding the precise degradation pathways from first life through postmortem analyses, in situ monitoring, and modeling are crucial for developing reliable second-life battery applications. Incorporating these degradation insights into battery selection and management protocols will enhance performance predictability and safety in second-life deployments.[88]

The technical viability of retired EV batteries in two second-life applications first one for power smoothing grid-scale PV plants and the second one for home demand response management, is covered in the article.[64] Batteries with various first-life aging histories and SoH were included in the analysis. The study

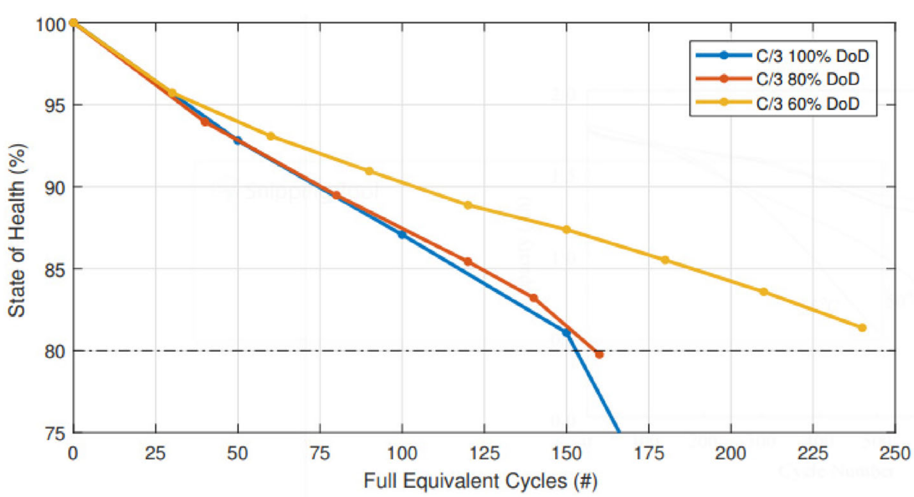

Figure 6. The influence of DoD on capacity retention over battery lifetime, where the black line indicates end of life (80% SoH). The graphic is reproduced with permission.[82] Copyright 2018, Energies.

Table 2. Summary of prior degradation effects on second-life performance.

Study	Chemistry	1st-life stressors	2nd-life outcome	Key insight
[64]	NMC/C	High DoD, 0 °C storage	Lower SoH, unstable cycles	Low-temperature cycling accelerates lithium plating
[89]	NMC	Moderate aging (≈80% SoH)	Predictable degradation	Batteries aged optimally perform best
[88]	LFP/NMC	Variable SoC range	Fast 2nd-life fade for severe Li plating	SoH alone insufficient to predict 2nd-life suitability

demonstrated that it is invalid to use 70–80% residual capacity as a retirement criterion for car batteries. Additionally, after reaching the aging knee, where the major aging process shifts, it was discovered that reusing batteries did not significantly slow down aging. Overall, the study found that the only batteries that would provide long-lasting second-life performance were those that were retired from automotive usage in the early stages of degeneration. Nevertheless, 800–1900 Cycles in automotive service still showed sufficient capability for second-life use.

Second-life performance is strongly influenced by the initial SoH. For instance, cells aged at optimal conditions (+25 °C) have a higher SoH at the beginning (75–80%) and perform better in their second life than cells aged under harsher conditions (0 °C), which have a lower SoH (70–75%). Cells cycled at 0 °C exhibit unpredictable behavior, while cells aged at +25 °C have a higher cycle durability. First-life conditions influence degradation mechanisms such as loss of active material (LAM) and lithium plating; cells with severe lithium plating at lower temperatures have a rapid capacity fade, whereas cells aged optimally last longer in their second-life. Additionally, cells aged at 0 °C have lower thermal stability, which raises safety concerns for second-life applications.[89]

Table 2 summarizes selected studies that explore the relationship between first-life stress factors and second-life performance outcomes for LIBs with different chemistries. The table highlights how specific conditions such as low-temperature cycling, depth of discharge, and SoC range impact subsequent degradation behavior. Notably, it demonstrates that second-life performance cannot always be predicted by SoH alone, emphasizing the importance of understanding prior stress history.

4. Conclusions

The introduction of second-life LIBs marks a critical step toward achieving sustainability and advancing the concepts of the circular economy within the energy sector. The aging mechanisms of these batteries have been reviewed in this work, along with the potential and challenges they bring in different applications. Important degradation processes such as electrolyte decomposition, cycling degradation, and capacity fading, highlight the need for ongoing innovation in battery design, maintenance, and management systems. Although the performance and dependability of second-life batteries have improved due to developments in diagnostics and battery management systems, there are still a number of important questions that need to be answered. These include how prior usage, heterogeneity, and operational conditions affect battery aging. Despite these challenges, second-life batteries offer a cost-effective and environmentally sustainable alternative to new batteries, especially for applications with lower performance demands. To realize their full potential, nevertheless, more research is needed to improve safety procedures, standardize testing procedures, and optimize aging mitigation tactics. In order to make sure that second-life batteries have more positive environmental effects than negative ones and to make it easier for them to be integrated into renewable energy systems, future work should concentrate on assessing the life cycle of these batteries. In the end, repurposed LIBs have the potential to be extremely important in lowering resource usage and fostering greener, more robust energy systems.

5. Future Directions of Focus

Based on the existing literature on the aging of second-life LIBs, several gaps, problems, and future directions can be identified, for example, heterogeneity of second-life batteries, since they come from diverse sources, including electric vehicles, consumer, prosumer and flexumer electronics, and grid storage systems. This heterogeneity introduces variability in battery chemistry, state of health, and degradation patterns.

Future research should consider how battery management systems (BMS) can be leveraged not only to mitigate degradation during second-life operation but also during first-life usage, where intelligent control strategies may reduce stress on cells and extend overall lifespan. Another possible research area could be standardized testing protocols; establishing uniform testing procedures and metrics would facilitate comparison between studies and enable more reliable evaluation of aging mechanisms and mitigation strategies. Also, LIBs have the potential to be more integrated with renewable energy systems by providing grid stabilization and energy-storage services. However, there are technical, economic, and regulatory barriers to their widespread adoption in this context. Future research should conduct comprehensive life cycle assessments to evaluate the environmental benefits and trade-offs associated with second-life battery utilization and recycling. Future work should pursue multi-modal aging studies, robust LCA modeling, and harmonized testing standards to support second-life battery integration into sustainable energy systems. This review highlights the need for a comprehensive framework that integrates chemical, thermal, and mechanical aging indicators. We recommend establishing standardized testing protocols across chemistries and applications, implementing holistic diagnostics through combined real-time BMS monitoring and laboratory characterization, enforcing safety assessments before and after repurposing, and incorporating circular economy metrics such as life-cycle assessment and cost-benefit analysis to evaluate second-life battery viability.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: aging mechanism · battery aging · degradation · electric vehicle · second-life batteries

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