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Review

Regeneration of Hybrid and Electric Vehicle Batteries: State-of-the-Art Review, Current Challenges, and Future Perspectives

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Abstract: Batteries have been integral components in modern vehicles, initially powering starter motors and ensuring stable electrical conditions in various vehicle systems and later in energy sources of drive electric motors. Over time, their significance has grown exponentially with the advent of features such as "Start & Stop" systems, micro hybridization, and kinetic energy regeneration. This trend culminated in the emergence of hybrid and electric vehicles, where batteries are the energy source of the electric traction motors. The evolution of storage for vehicles has been driven by the need for larger autonomy, a higher number of cycles, lower self-discharge rates, enhanced performance in extreme temperatures, and greater electrical power extraction capacity. As these technologies have advanced, so have they the methods for their disposal, recovery, and recycling. However, one critical aspect often overlooked is the potential for battery reuse once they reach the end of their useful life. For each battery technology, specific regeneration methods have been developed, aiming to restore the battery to its initial performance state or something very close to it. This focus on regeneration holds significant economic implications, particularly for vehicles where batteries represent a substantial share of the overall cost, such as hybrid and electric vehicles. This paper conducts a comprehensive review of battery technologies employed in vehicles from their inception to the present day. Special attention is given to identifying common failures within these technologies. Additionally, the scientific literature and existing patents addressing regeneration methods are explored, shedding light on the promising avenues for extending the life and performance of automotive batteries.

Keywords: electric vehicles; batteries; regeneration; reutilization; sustainability; review and perspective; life cycle

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1. Introduction

The latest report from the International Energy Agency, "World Energy Outlook 2023", highlights a notable threefold increase in electric vehicle (EV) sales over the past two years. Projections suggest that by 2030, electric vehicles will constitute 40% of the global vehicle fleet, reaching an estimated 1.4 billion vehicles worldwide, as reported by the European Automobile Manufacturers' Association (ACEA). This transformative shift towards electric mobility necessitates a thorough examination of its environmental impact and considerations for sustainable development. Prominent publications addressing the life-cycle analysis of electric and combustion vehicles [1–3] consistently report a significant reduction in emissions and pollutants favoring electric vehicles. Notably, these studies underscore the imperative for targeted research on the environmental impact of batteries—an integral component of the electric vehicle ecosystem. The scarcity and high value of metals crucial for battery production, coupled with potential pollution risks if not adequately managed at the end of their life cycle, highlight the importance of adopting a circular economy perspective.

According to the escalating number of decommissioned batteries from hybrid and electric vehicles and the anticipated surge by 2030, addressing their end-of-life management

becomes imperative. Approaching the issue from a circular-economy standpoint entails exploring alternatives like reuse in applications with lower demands, such as stationary energy storage for renewables and recycling. Notably, the circular perspective dismisses recovery as a viable solution. By considering the specific literature, Kotak et al. [4] underscored the superiority of battery reuse over recycling, positing that it delays recycling and allows time for market evolution towards innovative recycling technologies. Cusenza et al. [5] examined the potential integration of electric vehicle batteries into residential stationary applications, delaying the need for treating batteries as waste for approximately 10 years, albeit acknowledging the persistent challenge of battery waste. Canals et al. [6] discussed the advantages of reuse over recycling and delineated scenarios where 20% of retired batteries could find utility in low-demand applications, 70% in high-capacity applications, and 10% reintegrated into vehicles. Concurrently, Andersson and Rade [7] stressed the necessity of establishing a circular economy in battery manufacturing to ensure an adequate supply of metals. While reuse presents a promising avenue, traditional applications are often impractical due to diminished performance. Thus, alternative applications with lower demands, such as renewable energy storage or load smoothing in buildings, are sought. A less conventional approach to end-of-life management involves regeneration, defined as restoring a battery's capacities to its original or comparable state. Regeneration, if successful, doubles the battery's lifespan, potentially allowing for multiple regeneration cycles. If regeneration becomes unviable, the battery can be repurposed or recycled, contributing to a substantial extension of its life cycle and mitigating material usage and waste at the end of its operational life.

Under this framework, this paper conducts a comprehensive review of battery technologies employed in vehicles from their inception to the present day. Special attention is given to identifying common failures within these technologies. Additionally, the scientific literature and existing patents addressing regeneration methods are explored, shedding light on the promising avenues for extending the life and performance of automotive batteries. The rest of the paper is structured as follows: Section 2 describes the different types of batteries and their commercial applications, Section 3 discusses the battery regeneration technology, and Section 4 summarizes this review and the proposed technologies and solutions for regeneration purposes. Finally, conclusions are given in Section 5.

2. Types of Batteries and Vehicle Applications

The report "Electric Vehicle Battery Technologies: From Present State to Future Systems" [8] outlined a taxonomy encompassing six distinct battery technologies utilized in electric vehicles: lead–acid "Pb-acid", nickel–cadmium "Ni-Cd", nickel–metal hydride "NiMH", lithium-ion "Li-ion", lithium-ion polymer "LiPo", and sodium–nickel–chloride "NaNiCl". Conversely, 'A Review of Current Automotive Battery Technology and Future Prospects' [9] used a similar classification, albeit without distinguishing between lithium-ion "Li-ion" and lithium-ion polymer "LiPo", resulting in an aggregate of five technologies. This classification alignment was echoed in [10], further reinforcing the consensus. For the purposes of this document, a standardized categorization is adopted, aligning with the common classification from the cited works: Pb-acid, Ni-Cd, NiMH, and Li-ion, ensuring methodological uniformity in the examination of electric vehicle battery technologies. Table 1 summarizes and compares the main characteristics of such battery technologies identified and used in automotive vehicles.

In general, the high cost of batteries remains a significant barrier to widespread electric vehicle adoption. Innovations in manufacturing processes, material sourcing, and economies of scale are essential for making electric vehicles more affordable. The production of batteries relies on scarce resources such as lithium, cobalt, and nickel. Sustainable sourcing and recycling efforts are needed to ensure a stable supply chain and minimize environmental impact.

Table 1. Characteristic comparison of battery technologies used in automotive vehicles.

Technology	Nominal Voltage (V)	Energy Density (Wh/kg)	Cost (EUR/kWh)	Number of Recharging Cycles (Life Span)	Self-Discharge Coefficient (%/24 h)	Market Size in 2023 (USD B) [11]
Pb-acid	2	10–40	25–40	500	1	12.06
Ni-Cd	1.20	30–60	200–500	1350	5	-
NiMH	1.20	45–80	275–550	1350	2	1.61
Li-ion	3.25–3.60	55–200	400–800	1000	1	7.33

2.1. Lead–Acid Technology

The lead–acid technology, the first among those mentioned, has a longstanding history of application in traditional vehicles, where it serves to furnish the requisite energy for initiating the motor and sustaining electrical voltage within the vehicle. Following the vehicle's initiation, the alternator predominantly upholds voltage in the electrical circuits, yet the battery supplements during instances of low revolutions and heightened load. This battery type was the pioneering technology developed and remained unchallenged until the advent of Ni-Cd technology in the 1990s. Lead–acid batteries have also found utility in smaller vehicles aiding individuals with disabilities, golf carts, and industrial vehicles for material handling within industrial premises. Presently, this technology persists in use, notably in cost-effective electric vehicles like REVA [12], ALKE [13], or MELEX [14].

The chemical reaction transpiring within a lead–acid battery is articulated as follows:

$$PbO_2 + 2H_2SO_4 + 2e^- \rightarrow 2H_2O + PbSO_4 + SO_4^{2-}$$

$$Pb + SO_4^{2-} \rightarrow PbSO_4 + 2e^-$$

Battery depletion occurs when $PbSO_4$ crystallizes, and these lead crystals fail to initiate the reverse reaction, yielding Pb once more. Moreover, these lead crystals accumulate on the battery plates, resulting in an elevated electrical resistance. This impedes the smooth flow of electric current, concurrently diminishing the available Pb surface for the reaction. Lead–acid batteries have been a traditional choice for automotive applications, particularly in internal combustion engine vehicles. They are cost-effective but have lower energy density and a shorter lifespan compared to lithium-ion batteries [8,9].

2.2. Nickel–Cadmium Technology

Nickel–cadmium batteries have seen limited application in the automotive industry primarily due to their lower energy density, approximately half that of lead batteries. Another significant drawback is the presence of a memory effect, making it impractical to fully discharge the battery in a vehicle for recharging without rendering the vehicle inoperable for extended periods. This memory effect, extensively investigated in studies [15], mirrors a process akin to lead batteries where crystal formation occurs and deposits on active surfaces, diminishing the capacity to facilitate the reaction. With a prolonged presence of crystal generation, there's a risk of breaking the insulator separator, leading to self-discharge or a potential short circuit in the battery.

The chemical reaction occurring within a Ni-Cd battery is as follows:

$$\text{Oxidation reduction reaction}: \ Cd + 2OH^- \rightarrow Cd(OH)_2 + 2e^-$$

$$\text{Reduction reaction}: \ 2NiO(OH) + 2H_2O + 2e^- \rightarrow 2Ni(OH)_2 + 2OH^-$$

Only a handful of vehicles have adopted this battery type; notable examples include the *Renault Clio Electrique*, equipped with 19,100 Ah Ni-Cd batteries, and the *Peugeot 106* electric.

2.3. Nickel–Metal Hydride (NiMH) Technology

This technology stands out as one of the pioneering choices extensively employed in hybrid vehicles due to its absence of a memory effect, enhanced capacity, lower self-discharge rate, and a greater number of charge and discharge cycles. The Toyota Prius, marking the debut of hybrid vehicles, prominently featured this battery type, subsequently followed by various other brands adopting the technology, including *Toyota Rav4*, *Honda Insight*, *Honda Civic*, *Peugeot 3008*, and *Lexus RX*, among others. However, the landscape shifted with the widespread adoption of Li-ion technology in pure electric vehicles.

The chemical reaction occurring at the positive electrode is

$$Ni(OH)_2 + OH^- \rightarrow NiOOH + H_2O + e^-,$$

at the negative electrode, it is

$$M + H_2O + e^- \rightarrow MH + OH^-,$$

and in the fuel cell, it is

$$Ni(OH)_2 + M \rightarrow NiOOH + MH$$

Battery degradation is primarily induced by several factors, including elevated temperatures, overcharging, over-discharging, depth of discharge, and high-intensity charging and discharging. A comprehensive study [16] identifies nine key factors contributing to the loss of life in these batteries. Notably, the decline in capacity is attributed to the degradation of electrodes resulting from the oxidation of metal hydride (MH) alloys in the negative electrode. NiMH batteries are commonly used in hybrid vehicles. They have a higher energy density than lead–acid batteries but lower than lithium-ion. NiMH batteries are known for their reliability and safety, but they are gradually being replaced by lithium-ion technology.

2.4. Li-Ion Technology

Li-ion batteries currently dominate the landscape of pure electric vehicles, surpassing NiMH batteries, and have not yet been commercially displaced by any alternative technology in electric vehicles. Distinguished by their absence of a memory effect and lower self-discharge compared to Ni-based batteries, Li-ion batteries stand as the preferred choice. The electrochemical process during the charging phase is as follows:

The Li-ion battery anode cell is governed by

$$Li_x + C_6 + xLi^+ + xe^- \rightarrow LiC_6,$$

and the cathode cell by

$$LiCoO_2 \rightarrow xLi + xe^- + Li(1-x)CoO_2.$$

During the discharge process, the anode cell is governed by

$$LiC_6 \rightarrow Li_xC_6 + xLi^+ + xe^-,$$

and the cathode expression is

$$xLi + xe^- + Li(1-x)CoO_2 \rightarrow LiCoO_2.$$

The electrochemical processes within the cells that contribute to aging involve the formation of the solid–electrolyte interphase (SEI) in the anode. While crucial for charging and discharging capacity, the SEI must remain stable over time and facilitate lithium-ion conduction. The growth of this layer can even lead to detachment from the anode, further increasing impedance by losing contact with the copper layer responsible for electron

collection and electric current formation. Additionally, corrosion of lithium in the anode is identified as another aging mechanism, resulting in the depletion of available lithium ions and, consequently, a decrease in capacity; see Figure 1. According to the specific literature, Xiong et al. [17] conducted an exhaustive examination of lithium-ion battery aging methods, concluding that the aging processes can be categorized into capacity reduction, increased internal resistance, and internal breakdown of the battery. These effects are instigated by physical and chemical reactions within the battery, resulting in the loss of lithium or active material, primarily influenced by different temperatures and charging/discharging speeds. Collath et al. [18] acknowledged the same aging mechanisms but offered an alternative classification based on whether the effects manifest in the anode or the cathode and highlighted that the main cause indicated in the anode is the growth of the SEI layer due to the reaction of graphite with lithium, which causes loss of lithium and an increase in internal resistance; in the cathode, a mechanism similar to that of the anode occurs, forming a thinner layer than the SEI, but that also increases the internal resistance. In a similar way, Heiskanen et al. [19] affirmed that aging transpires as the SEI expands due to reactions in the electrolyte, avoiding any ion diffusion and escalating internal resistance.

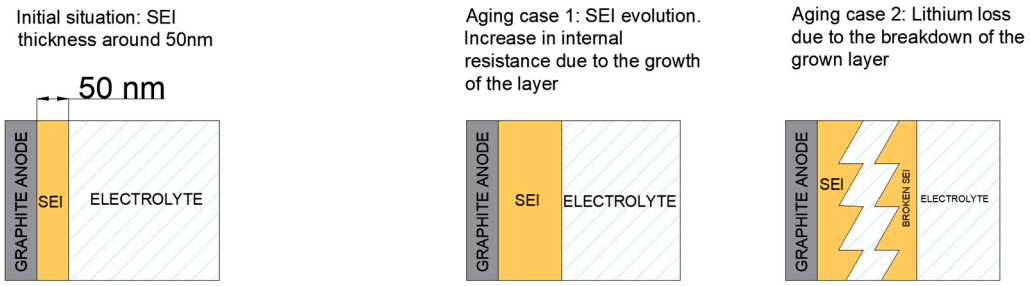

Figure 1. Initial situation of the SEI and main aging factors.

For a more complete aging analysis, additional degradation mechanisms must be considered. In this way, Jiang et al. [20] reviewed different degradation processes taking place at the cathode, anode, and electrolyte in nickel-rich cathode-based lithium-ion batteries. Recently, Mayemba et al. [21] affirmed that the aging of a Li-ion battery was influenced by various parameters, including the charge and discharge profiles, the state-of-charge (SOC) window within which the tests are conducted, and the temperature. Han et al. [22] provided an extensive examination of the primary concerns regarding battery degradation throughout its entire life cycle and considering various anode and cathode materials.

In summary, it can be affirmed that, nowadays, lithium-ion batteries are widely used in a variety of applications, including electric vehicles (EVs), smartphones, and laptops, among others. In general, they offer relevant characteristics, such as high energy density, long cycle life, and relatively low self-discharge rates. However, concerns about resource availability, safety, and cost persist.

3. Battery Regeneration Technologies

Regeneration is conceptualized as the restoration of a battery's capacities to either its original state or a state closely resembling the original. Specific characteristics required for a normal performance are reinstated for an expected life similar to a new battery. By considering the specific literature, for example, [23], two methods for battery regeneration can be identified: (i) The first group involves a chemical process wherein reactions are used to revert the anode, cathode, and electrolyte to their original state before the battery experienced degradation. This method requires access to the inner of the cells for the reagents in order to react with the deteriorated components. However, contemporary cells in hybrid and electric vehicles are typically highly encapsulated, mainly for safety requirements, preventing access to damaged parts for chemical reactions without causing destruction. This encapsulation avoids reactions with O_2 and protects against physical damage, accidents, or unauthorized handling, making it difficult to implement the corresponding

chemical regeneration under these circumstances. (ii) The alternative regeneration method involves a series of electrical and physical processes, achieving effects such as temperature changes and the application of different electric currents with varying waveforms and pulses. Unlike the chemical approach, this solution does not necessitate opening the cell, allowing for regeneration without the need to remove the battery from the vehicle.

Within the context of a circular economy, this review also encompasses a discussion on battery recycling. As detailed in the subsequent sections, the regeneration techniques do not delineate the state of health (SOH) at which regeneration is viable and when it is not. Consequently, it is always essential to carry out battery regeneration attempts initially. Therefore, in cases where the regeneration process does not achieve a SOC suitable for the intended application, a transition to recycling becomes imperative. The objective is then to recover the battery's components for subsequent utilization in the production of new components.

3.1. Regeneration Systems in Lead–Acid Batteries

In the year 2000, the European patent titled "Method of Regenerating Lead Storage Batteries" [24] was published, outlining a lead–acid battery regeneration method. The approach involves applying current pulses with a frequency of around 10 kHz from the positive electrode to the negative electrode. This aims to reduce the lead sulfate deposited on the electrode's surface. A subsequent activation step for the positive electrode involves electrochemical doping, where direct current is applied to the battery with a carbon suspension acting as the electrolyte. The carbon suspension is obtained through the electrolytic oxidation of the positive carbon electrode in a water system. This method not only reduces lead sulfate but also activates the positive electrode through electrochemical doping.

In 2013, the Spanish patent "Regeneration battery method for lead-acid technology" (in Spanish) was published, proposing a method for regenerating spent lead batteries [25]. This method involves applying a sequence of direct current pulses at different frequencies, ranging from 15 kHz to 25 kHz, between the battery electrodes. This process breaks the lead sulfate crystals formed on the electrodes. The method includes stages of charging and discharging, as well as a battery state analysis stage. Importantly, no additional chemical compounds are required, distinguishing it from earlier methods. The absence of an electrolyte for regeneration makes this method an advancement in lead–acid battery regeneration. Commercially, this method is implemented through regeneration systems produced by various manufacturers, including Maroo [26], Macbatec [27], Amperis [28], Prime [29], BRT [30], Battmaster [31], and DF48 [32]. Widely accepted and commercially successful, this method has been studied in [33], comparing the physical–chemical state of electrodes in regenerated and unregenerated batteries. The study concluded that in the regenerated system, $PBSO_4$ crystals are not visible, indicating their elimination in the regeneration process. Table 2 summarizes the lead–acid battery regeneration processes. The destructive testing (DT) parameter is also included in Table 2 and in the rest of analysis, giving additional information regarding the process in terms of breaking (or not) the cell. Regarding lead–acid technology, it can be affirmed that regeneration has evolved into a mature, profitable, and commercially established process.

Table 2. Lead–acid battery regeneration process summary.

Ref.	Published by	Year of Publication	Process	Physical Method	Chemical Method	Commercial Use	Destructive Testing (DT)
[24]	Patent	2000	Regeneration	Current pulses	Carbon electrolyte addition	No	No
[25]	Patent	2013	Regeneration	Current pulses		Yes	No
[33]	Journal paper	2013	Regeneration	Current pulses		Yes	No

3.2. Regeneration Systems in Ni-Cd Batteries

There is a lack of contributions in the specific literature focused on the regeneration of Ni-Cd batteries, although recycling experiences for this technology have been documented, incorporating processes in conjunction with other technologies discussed in this review. The authors only identified [34] as a Ni-Cd-specific publication for battery regeneration processes, which described a method for regenerating Ni-Cd batteries using high-intensity pulses (ranging from 100 A to 300 A) to disrupt the crystalline layer of amorphous nickel hydroxide. Through this approach, they achieved a 41% recovery in battery capacity. Notably, the procedure bears similarities to the one described earlier for lead–acid batteries. Unfortunately, details regarding pulse shape, duration, or frequency are not provided, leaving uncertainty as to their relevance or potential complications in reproducing the experiment. A future avenue of research could be the application of this regeneration method and the verification of its operation or its discarding. In [35], the authors carried out several battery charging experiments with current pulses which indicated the value of the peaks and the frequency, after which they compared the electrodes using microscopy and concluded that the morphology in the electrodes using a conventional system and another pulse was similar and that the discharge capacity of the cell in both cases was the same.

Regarding the regeneration efforts of nickel–cadmium "NiCd" batteries, in conclusion, it appears that the available methodologies can be broadly categorized into two groups. On the one hand, certain methods lack clarity regarding the explicit steps required to replicate the regeneration process. On the other hand, some approaches provide procedural guidelines but ultimately assert that the outcomes closely resemble those achievable without undergoing the regeneration process. Consequently, it appears that regenerating this particular type of battery may not be a feasible endeavor.

3.3. Regeneration Systems in NiMH Batteries

Contributions are categorized into three groups: (i) regenerating the entire battery without dismantling it, (ii) regenerating materials to reshape cells, and (iii) material recycling for alternative applications.

3.3.1. Material Regeneration Processes

Nan et al. [36] outlined a recycling process utilizing a specialized machine that involves crushing and subsequent treatment with acid. This results in a solution containing the necessary materials for manufacturing new battery cells. In contrast, Wang et al. [37] detailed a method that involves breaking the battery, extracting the alloy, and regenerating it through a simple chemical process with the addition of a mild acid. The cell is then reformed by melting in an inert atmosphere. Additionally, Li et al. [38] explained a technique to regenerate the cathode by extracting its materials through physical methods such as crushing and pulverizing. Following this, Li_2CO_3 is added, and a high-temperature treatment in an inert atmosphere is applied. Using this recycled material, cells are remanufactured with an efficiency of 97.77%. In summary, the contributions suggest that regenerating materials in NiMH cells involves transforming them into powder through a physical method, treating them with acid, and reshaping the electrodes through thermal treatment in an inert atmosphere.

3.3.2. Material Recycling Processes

Certain recycling methods exclusively use mechanical processes. Mir et al. [39] investigated the effects of temperature and microwave treatment as a pretreatment for recycling to decompose materials constituting the electrodes, performing several experiments at different temperatures between 200 °C and 1000 °C, as well as different times and exposure to microwaves with a power of 900 W between 5 and 20 min. The results were better in microwave heating for the decomposition of materials. Tsunekawa et al. [40] explained a process that involves the separation of metallic components of different sizes after con-

verting them to powder using magnetic fields. From the results, the process was suitable for the separation of the anode and cathode activation agents and their substrates. Shin et al. [41] used a method for element separation through thermal treatments, allowing heating between 300 °C and 600 °C for 2 h. After this initial separated treatment, a powder was obtained with all components prepared for the next recycling step through hydrometallurgy. Bertuol et al. [42] proposed using milling and magnetic effects to separate the magnetic part of batteries, obtaining very good results through two grinding stages and three magnetic separation stages, losing only 10% of the weight of the initial material in the process. Tenório et al. [43] utilized mechanical means and magnetic fields to extract metals, very similar to the previous process. Müller et al. [44] established a recycling method solely relying on mechanical processes. In addition to NiMH, Huang et al. [45] also incorporated Ni-Cd using a mechanical, magnetic, gravity, and air- and vacuum-current-based approach. Figure 2 shows a general scheme of the NiMH battery recycling process.

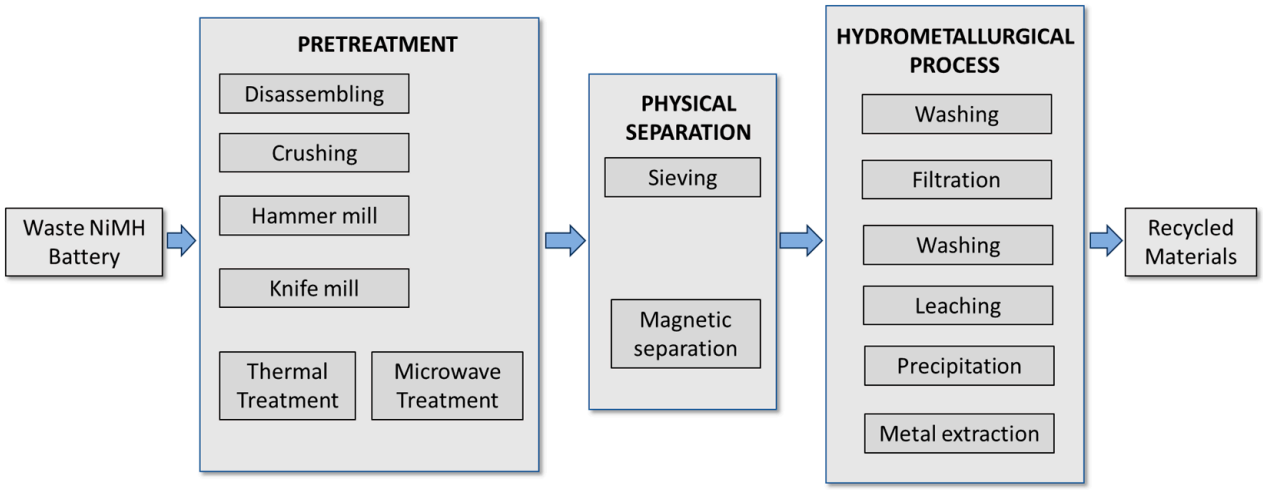

Figure 2. Diagram of the general recycling process in NiMH batteries.

Commercial automotive NiMH batteries include lanthanum; the cathode is a mixture of metal hydrides like lanthanum hydride. The active material in the cathode is hydrogen, stored in the metal hydride structure. Depending on its composition, it can contain between 1% and 7% hydrogen by weight. As an example, a Toyota Prius battery requires approximately 10 kg of lanthanum, being the cerium also used to store hydrogen. Most methods employed for recovering rare earths, such as nickel and cobalt, share similar principles with slight variations. Generally, these methods involve opening batteries to extract electrodes, mechanically pulverizing them, and treating them with various acids, primarily H_2SO_4 and NaOH. Examples can be found in [46], focused on the extraction of La and Ce, carrying out processes with different concentrations of sulfuric acid and temperatures, obtaining satisfactory results in all cases. Zhang et al. [47] used HCl and established the most favorable concentration and temperature conditions for this process. Pietrelli et al. [48] corroborated the process, ensuring the recovery of 80% of the rare earths. Other authors carried out the same process establishing the portents of rare earths recovered [49–51], varying these between 80% for La and 99% for Sm. Santos et al. [52] described a method also applicable to Li-ion and Ni-Cd technologies, yielding metals of high purity.

There are some variants, such as the study conducted by Nayl [53], which used HCl and HNO_3 for metal extraction. Provazy et al. [54] used the same technique not only for NiMH but also for Ni-Cd and Li-ion batteries, concluding with an organic solvent for metal extraction. Kavozy et al. [55] concluded the method with CH_4 dry reforming. Other methods focus on metal extraction using organic extractants after mechanically pulverizing the electrodes. Granata et al. [56] employed a mixture of NiMH and Li-ion batteries [57,58]. A variation involves hydrochloric acid treatment, as seen in [59,60], with [61] using an organic extractant post-HCl treatment; alternatively, [62] used two

organic extractants, D2EHPA and Cyanex, for the extraction of cobalt and nickel. The same method is used in [63]; however, the process ends with the preparation of crystallized $Ni(OH)_2$. Reference [64] carried out the same process, obtaining the concentrations of D2EPHA and Cyanex that maximize the extraction of cobalt and nickel. In [65], the authors performed the same procedure with Cyanex but focused their study on the optimal concentrations for the extraction of La and Nd. Reference [66] used the same method and established what the order of extraction of the metals should be. Additionally, [67] used the same method, emphasizing that the method meets the circular economy criteria by being able to recover organic agents. A similar method was proposed by Petranikova et al. [68], developing it for a pilot plant and optimizing its use in the plant. According to the specific literature, the recycling methods for NiMH batteries have been widely explored and present various approaches. However, a prevalent method involves converting electrodes into powder through mechanical means and treating them with H_2SO_4 and NaOH, resulting in a solution where metals are dissolved and subsequently separated using branded organic extractants, mainly the commercial brands D2EPHA and CYANEX.

In addition to the hydrometallurgical method, other recycling methods can be found in the specific literature. As an example, the pyrometallurgical method [69], where high purity alloys for new batteries are obtained using vacuum furnaces in hydrogen gas at temperatures until 900 °C. Another approach can be found in [70], in which the temperature reached 1500 °C, and the process could obtain high-purity rare earths, up to 96% purity, using magnetic separation. Recently, Li et al. [23] proposed the electrochemical approach for recycling spent Li-ion batteries by substituting traditional chemical leaching with applied current as the driving force for reactions. This approach presents significant potential to enhance recycling efficiency, minimize reagent usage, and mitigate environmental risks.

3.3.3. Battery Regeneration Discussion

There are scarce methods for regenerating this type of battery, with only three relevant patents identified. In 1997, it was published a patent outlining a method to reactivate electrodes by washing the electrolyte with acids and bases, subjecting the cells to electric currents at different temperatures [71]. In 2014, the Spanish patent "Method for regenerating NiMH batteries" [72] detailed a controlled discharge at constant intensity in two different steps and a controlled charge at constant intensity to regenerate a battery composed of several cells. In 2021, the patent "Method for reconditioning NiMH battery cells" suggested restoring internal resistance by introducing oxygen into the cell [73]. Additionally, Li et al. [74] highlighted improved electrochemical performance through ultrasound-induced detachment of the oxidation layer on the negative electrode, though the work lacks details on replicating the method. However, there is no literature on practical cases following these patents, and no market for NiMH battery regeneration has emerged. In the opinion of the authors, this absence raises doubts about the efficacy of these methods.

Future research should go along the lines of testing these patents to check if they really achieve the regeneration of NiMH batteries. In [75], it is discussed how to charge batteries in a more efficient way by controlled current pulses, establishing the width, duration, and intensity of the pulses to achieve a decrease in the internal pressure of the cell, less oxidation in the electrodes and better conservation of their crystalline structure. This method is not a regeneration system, but it helps to maintain the cell or improve its properties lost through use. Chen et al. [76] provided a way of charging useful to revive the spent batteries. It consisted of charging through a sine wave. In the contribution, the authors gave both the voltage amplitude and the frequency values in detail. According to this method, it was possible to recover batteries damaged by above 80% SOH. After a study of the battery resistance in the spectrum, the curve obtained was very similar to a new battery, providing the suitability of the method for the recovery of batteries. Table 3 shows a NiMH Battery regeneration process summary.

Table 3. NiMH battery regeneration process summary.

Ref.	Published by	Year of Publication	Process	Physical Method	Chemical Method	Destructive Testing (DT)
[47]	Paper	1998	Recycling	Mechanical	H_2SO_4, Ammonia, Oxalic acid	Yes
[62]	Paper	1999	Recycling	Mechanical	H_2SO_4, Ammonia, Oxalic acid	Yes
[39]	Paper	2021	Recycling	Temperature and microwave	Acid treatment	Yes
[48]	Paper	2002	Recycling	Mechanical	H_2SO_4	Yes
[37]	Paper	2002	Material Regeneration	Ultrasound, smelting	Weak acid, Ar	Yes
[43]	Paper	2002	Recycling	Mechanical, Magnetic		Yes
[75]	Paper	2004	Regeneration	Current pulses		No
[74]	Paper	2005	Regeneration	Ultrasound		No
[42]	Paper	2006	Recycling	Mechanical, magnetic		Yes
[44]	Paper	2006	Recycling	Mechanical		Yes
[40]	Paper	2007	Recycling	Magnetic field		Yes
[50]	Paper	2008	Recycling	Mechanical	H_2SO_4, other acids	Yes
[63]	Paper	2009	Recycling	Mechanical	H_2SO_4, NaOH	Yes
[55]	Paper	2009	Recycling	Mechanical	H_2SO_4, HCL, HNO_3	Yes
[64]	Paper	2010	Recycling	Mechanical	H_2SO_4, NaOH	Yes
[53]	Paper	2010	Recycling	Mechanical	H_2SO_4, NaOH, HCl, HNO_3	Yes
[45]	Paper	2011	Recycling	Mechanical, magnetic, air vacuum		Yes
[54]	Paper	2011	Recycling	Mechanical	H_2SO_4, NaOH	Yes
[46]	Paper	2012	Recycling	Mechanical	H_2SO_4, NaOH	Yes
[66]	Paper	2012	Recycling	Mechanical	H_2SO_4, NaOH, Organic Extractants	Yes
[57]	Paper	2012	Recycling	Mechanical	Organic Extractant	Yes
[52]	Paper	2012	Recycling	Mechanical	H_2SO_4, NaOH	Yes
[49]	Paper	2012	Recycling	Mechanical	H_2SO_4, other acids	Yes
[56]	Paper	2012	Recycling	Mechanical	Organic Solvents	Yes
[58]	Paper	2013	Recycling	Mechanical	Organic Extractant	Yes
[65]	Paper	2013	Recycling	Mechanical	H_2SO_4, NaOH	Yes
[72]	Patent	2014	Regeneration	Current pulses		No
[60]	Paper	2014	Recycling	Mechanical	Hydrochloric	Yes
[70]	Paper	2014	Recycling	Temperature		Yes
[41]	Paper	2015	Recycling	Temperature		Yes
[69]	Paper	2015	Recycling	Temperature		Yes
[61]	Paper	2015	Recycling	Mechanical	HCl, Organic Extractant	Yes
[68]	Paper	2017	Recycling	Mechanical	HCl, Organic extractants	Yes
[51]	Paper	2017	Recycling	Mechanical	H_2SO_4, NaOH	Yes
[76]	Paper	2019	Regeneration	Current pulses		No
[67]	Paper	2020	Recycling	Mechanical	H_2SO_4, Organic Extractants	Yes
[73]	Patent	2021	Regeneration		O_2 adding	Yes

3.4. Regeneration Systems in Li-Ion Batteries

In this technology, two types of processes have been found: (i) based on regenerating the materials from which spent cells are made to produce new cells, and (ii) recycling the materials for alternative use. Novel eco-friendly regeneration technologies can be found in [77]. The authors affirmed that formulating a fully recyclable and regenerative Li-ion

battery stands out as the most auspicious approach to address the issue of such batteries. Moreover, Wang et al. [78] concluded that it is imperative to develop repurposing strategies, which may encompass various options such as recycling or transitioning to a second life in stationary energy storage applications. The authors also suggested that non-destructive methods are dependable for identifying active aging mechanisms.

3.4.1. Material Regeneration Processes

Zhang et al. [79] introduced a method for regenerating battery anode material after it reaches the end of its life. This approach involves a thermal treatment followed by a chemical treatment, successfully regenerating the anodes, albeit with a lower performance compared to new material. Yang et al. [80] presented a method for recycling $FePo_4$, obtaining material in a crystalline form; the process consisted of treating the waste with HCl and H_3PO_4 for 1–4 h at a temperature between 80 and 110 °C, then cleaning and filtering and burning at 600 °C, thus forming regenerated hexagonal $FePO_4$ crystals. Li et al. [81] addressed cathode material regeneration by opening cells and treating them with a NaOH solution, achieving over 90% performance. Zhang et al. [82] explored regenerating spent $LiCoO_2$ batteries through a chemical process, achieving a 97% regeneration of lithium and producing high-efficiency $LiCoO_2$ cathodes. This process involves opening cells and extracting material for chemical treatment. Ji et al. [83] studied lithium recovery and $LiFePo_4$ battery regeneration using an organic lithium salt, emphasizing its effectiveness but requiring a chemical reaction that necessitates disassembling and reassembling the battery. The study concluded that this regeneration method is economically more viable than other recycling systems. Liang et al. [84] presented a practical case of recycling used $LiFePo_4$, involving disassembling the battery, thermally treating the cathode, and reintroducing necessary elements to achieve the initial molar fraction, ultimately obtaining $LiFePO_4$ in its original state. Su et al. [85] successfully resynthesized spent $LiFePO_4$ through thermal treatments and mechanical processes without chemical treatment. Xu et al. [86] proposed a method where, after collecting cathode material in powder form, adding lithium returns the material to its original state. Tang et al. [87] outlined a method to recycle spent $LiFePO_4$ by extracting cathode material with chemical treatment and adding Li_2CO_3 and other agents to regenerate $LiFePO_4$ for new cells. Palaniyandy et al. [88] established a system to regenerate the cathode of $LiMn_2O_4$ cells through chemical and physical means, such as ultrasound, followed by thermal treatment and recrystallization. Meng et al. [89] regenerated the cathode of a battery through screening, Li_2CO_3 addition, and joint calcination to obtain regenerated cathode material. Fab et al. [90] reconstructed the cathode through extraction, lithium doping, and subsequent thermal treatment. Yue et al. [91] regenerated the cathode of cells through a simple thermal treatment, requiring cathode material extraction and cell destruction. Reference [86] obtained recycled $LiFePO_4$ by re-lithiation treatment in a Li-containing aqueous solution with controlled temperature and time and compared different methods of recycling like pyrometallurgical, hydrometallurgical, and direct recycling with the help of a life-cycle analysis and an economic analysis concluding that this method is greener and more efficient than others studied.

Methods focused on recycling graphite can also be found in the specific literature. Moradi et al. [92] explained the main methods that exist for this, concluding that the pyro and hydro methods fail to achieve a type of recycled graphite that contains the physical and chemical characteristics required for the manufacture of new batteries, so the best alternative is recycling through direct extraction. Liu et al. [93] concluded in a similar way, recycling graphite using a hydro method. Nevertheless, it is not valid for the requirements of new lithium batteries, but it is valid for sodium batteries. Yi et al. [94] studied the methods for recycling graphite, concluding in a similar way to previous authors: recycled graphite may not have the requirements for new lithium batteries but is valid for other types of batteries, such as sodium ones. Natarajan et al. [95] described the method to recycle the separator. They focused on the manufacture of a new battery, obtaining a satisfactory result due to the mechanical characteristics being similar to the new ones. Ghasemkhani et al. [96]

recycled the separator and managed to manufacture other types of plastics suitable for use in the manufacture of corrugated pipes. Dissolution techniques with organic solvents or the application of supercritical CO_2 are proposed for electrolyte extraction purposes, as was explained in [97]. While the methods for material regeneration vary, many involve converting cathodes into powder, adding the spent element, and thermally treating them at temperatures exceeding 800 °C to recrystallize and make them suitable for manufacturing new cells.

3.4.2. Material Recycling Processes

Yi et al. [98] outlined a straightforward approach to recover graphite from batteries with a purity exceeding 99.5%; the process consisted of dismantling the cell, heat treatment at 1673 K for 4 h in N_2, and separation of the graphite by ultrasonic vibration and sieving. Zhou et al. [99] introduced a system for reclaiming spent cathode material, achieving a recovery of 98.13% for lithium and 98.86% for cobalt. Nie et al. [100] explored a method for recovering cathode materials from $LiMn_2O_4$ batteries and repurposing them in the production of sodium-ion batteries, thereby utilizing spent batteries for a new generation of energy storage, [101] exposed an industrial and scalable system to extract all the elements of which the lithium battery is composed using techniques set forth in this document, with this method, it is possible to extract the electrolyte, the electrodes and traces of metal without the use of organic extractants. The patent [102] established a method for obtaining battery metals through thermal processes. Some researchers have experimented with metal extraction using bacteria, as demonstrated by [103–106], yielding satisfactory results that could present economic competition with purely chemical methods. The patent [107] showed how to obtain LiOH from waste Li batteries by lowering its temperature, controlling the PH with water, and adding LiOH; the resulting salts were refined to obtain LiOH or $LiCO_3$ with the addition of CO_2. Patent [108] used a method to mix the material of the positive electrode with manganese to obtain a mixture of Li ions/manganese ions and then burned it at 250 °C for 5 h to be filtered to obtain a Li-rich solution.

The main Li recycling scenarios included pyrometallurgical (Pyro), hydrometallurgical (Hydro), and direct recycling. By considering the specific literature, not all the materials could be collected by such techniques, and some of them are not economically profitable. Subsequently, every extracted material is in the purity grade to produce batteries again, or it can be refined to obtain it, with different profitability ratios [109]. Life-cycle analysis and environmental analysis show how GHGs are reduced with the main techniques of recycling; see Table 4 [110–112], where NC denotes not collected, EP denotes economically profitable, and EU denotes economically unprofitable.

Table 4. Main recycling scenarios, materials recycled, economics, and GHG reduction.

Technique	Li	Co	Ni	Mn	Al	Cu	Graphite	Si	GHG Reduction
Pyro	EU	EP	EP	EU	NC	EP	NC	EU	43
Hydro	EU	EP	EP	EP	EU	EP	EU	EU	90
Direct	EP	EP	EP	EP	EU	EP	EU	EU	16

4. Discussion

Tables 2–5 illustrate the reviewed studies on battery regeneration, categorizing them based on the type of technology investigated, specifying whether they are publications or patents, and identifying whether they address cell regeneration, material regeneration, or recycling. Additionally, it outlines the primary physical and chemical methods employed, indicates whether cell disassembly is necessary, and notes if a commercial application has been identified.

Table 5. Li-ion battery regeneration process summary.

Ref.	Published by	Year of Publication	Process	Physical Method	Chemical Method	Destructive Testing (DT)
[107]	Patent	1998	Material Regeneration	Cryogenic	LiOH addition	Yes
[103]	Paper	2008	Recycling		Bacteria	Yes
[105]	Paper	2009	Recycling		Bacteria	Yes
[54]	Paper	2011	Recycling	Mechanical	H_2SO_4, NaOH, organic solvents	
[52]	Paper	2012	Recycling	Mechanical	H_2SO_4	Yes
[104]	Paper	2012	Recycling		Bacteria	Yes
[106]	Paper	2014	Recycling		Bacteria	Yes
[81]	Paper	2017	Material Regeneration		NaOH solution	Yes
[79]	Paper	2018	Regeneration	Heat	Phenolic resin–ethanol solution	Yes
[38]	Paper	2019	Material Regeneration	Temperature	Li_2CO_3 addition	Yes
[100]	Paper	2019	Recycling	Temperature		Yes
[89]	Paper	2019	Material Regeneration	Temperature	Li_2CO_3 addition	Yes
[91]	Paper	2020	Material Regeneration	Temperature		Yes
[88]	Paper	2020	Material Regeneration	Ultrasound temperature		Yes
[87]	Paper	2020	Material Regeneration	Temperature	DMC, NMP, LI_2CO_3 addition, glucose addition, $Cu(NO_3)_2$ addition	Yes
[86]	Paper	2020	Material Regeneration	Temperature	DMC, NMP, Li addition	Yes
[98]	Paper	2020	Recycling	Ultrasound	Nitrogen	Yes
[85]	Paper	2020	Material Regeneration	Temperature		Yes
[84]	Paper	2020	Recycling	Temperature	NaOH solution Li, Fe, P addition	Yes
[86]	Paper	2020	Material Regeneration	Re-lithiation		Yes
[101]	Paper	2020	Recycling	Temperature	Li addition	Yes
[93]	Paper	2020	Material regeneration	Temperature	H_2SO_4, HCl	Yes
[90]	Paper	2021	Material Regeneration	Temperature	Li addition	Yes
[108]	Patent	2021	Recycling	Temperature	Manganese	Yes
[99]	Paper	2021	Recycling	Ultrasound	Malic acid, DL, H_2O_2	Yes
[80]	Paper	2022	Recycling		Several	Yes
[82]	Paper	2023	Regeneration		Several	Yes
[83]	Paper	2023	Regeneration		Organic Lithium salt	Yes

Note that this analysis delves into the regeneration methods for diverse battery types, such as lead–acid, nickel–cadmium (Ni-Cd), nickel–metal hydride (NiMH), lithium-ion (Li-ion), and other emerging technologies. One focal point of discussion revolves around chemical regeneration methods that involve the application of reagents to restore the anode, cathode, and electrolyte components to their original or near-original state. This chemical approach has shown efficacy but is hindered by the encapsulation of cells in modern electric vehicles, limiting access to deteriorated parts. Contrastingly, electrical and physical regeneration processes have been explored as an alternative avenue. These methods subject cells to temperature variations, diverse current types, intensities, and pulses. Although most publications focused on battery regeneration methods discuss and describe each corresponding method, only for lead-acid batteries is it explicitly explained in detail what potential effects the regeneration process should address inside the cell. However, regarding NiMH batteries, the authors commonly provide global results, excluding, in general, the processes carried out inside the cells by the specific regeneration processes. Unlike chemical processes, these approaches do not necessitate cell disassembly, potentially allowing regeneration without removing the battery from the vehicle. This could be a valuable advantage, especially in the context of electric vehicles with highly encapsulated battery systems. Furthermore, the discussion extends to the challenges associated with battery regeneration, including the limitations imposed by high temperatures, overcharging, overdischarging, and other factors leading to degradation. Understanding the causes of

degradation is crucial in devising effective regeneration strategies and enhancing the overall sustainability of batteries.

In the context of Li-ion batteries, a predominant technology in electric vehicles, the discussion explores the aging mechanisms that contribute to a decrease in capacity, an increase in internal resistance, and internal breakdown. Studies suggest that these aging processes are influenced by physical and chemical reactions, emphasizing the importance of comprehending the intricate mechanisms governing lithium-ion battery performance. The conversation also addresses the economic and environmental implications of battery regeneration compared to recycling. While regeneration can potentially double the lifespan of a battery and reduce the need for raw materials, recycling remains a critical aspect, especially when regeneration is no longer viable. Recycling methods vary, employing mechanical, chemical, or a combination of both processes to extract valuable materials to produce new batteries. In conclusion, the discussion on battery regeneration processes navigates through the complexities of chemical, electrical, and physical methods, considering the specific challenges posed by different battery technologies. It emphasizes the need for comprehensive strategies to address degradation, maximize lifespan, and contribute to the circular economy in the realm of energy storage. Velázquez et al. [113] analyzed Li-ion recycling technologies through the lens of circular economy principles. As such, the discussion was centered on the capacity of each technology to recover all components within Li-ion batteries. Nevertheless, significant obstacles, such as the heterogeneity of cell design and battery chemistries, must be overcome to remanufacture and reuse the cell components and, subsequently, to capitalize on promising opportunities [114].

The future challenges in battery regeneration processes encompass several critical aspects. Firstly, there is a need for the development of comprehensive and standardized regeneration protocols for various battery technologies, especially for nickel–cadmium (Ni-Cd), nickel–metal hydride (NiMH), and lithium-ion (Li-ion) batteries. Current gaps in knowledge and practical applications hinder the widespread adoption of regeneration methods beyond lead–acid technology. Additionally, research must focus on enhancing the efficiency of regeneration processes, ensuring that regenerated batteries meet or exceed the performance standards of new batteries. This includes addressing issues related to capacity, cycle life, and safety. Achieving a balance between economic viability and environmental sustainability is crucial, requiring advancements in cost-effective regeneration techniques that minimize environmental impact. Furthermore, the development of reliable and scalable commercial solutions for battery regeneration remains a challenge. Establishing standardized methodologies that can be applied across different battery types and brands is essential for creating a viable market for regeneration services. This involves collaboration between researchers, industry stakeholders, and regulatory bodies to set guidelines and standards. Addressing the limitations associated with breaking down encapsulated battery cells is another challenge. Many modern batteries, particularly those in electric vehicles, are designed with safety features that make accessing internal components difficult. Overcoming these challenges requires innovative approaches to safely and efficiently regenerate batteries without compromising safety measures. Lastly, the integration of circular economy principles into battery design and manufacturing processes will play a crucial role in addressing the environmental impact of battery waste. Developing batteries with easier disassembly and regeneration capabilities can contribute to a more sustainable life cycle.

5. Conclusions

Among the analyzed battery regeneration systems for four different technologies, it is concluded that lead–acid battery regeneration systems are well-established and widely used commercially worldwide, demonstrating successful outcomes across numerous brands. However, the regeneration systems for the other technologies are either underdeveloped or require further investigation. Specifically, the regeneration system for nickel–cadmium cells mirrors that of lead–acid, with the available publication lacking the necessary parameters

for replication. In the case of NiMH batteries, existing patents outline the process, but there is a dearth of commercial development or publications verifying the effectiveness of this method. Regarding Li-ion batteries, publications predominantly focus on regenerating materials for cell manufacture rather than regenerating the entire battery for direct use without industrial transformation.

Consequently, excluding lead–acid technology, where regeneration is well-established commercially, achieving practical regeneration of batteries from other technologies remains elusive. Future research should prioritize testing the efficacy of the cited patents on Ni-Cd and NiMH technologies and evaluating their performance to ascertain potential economic viability. The future challenges in battery regeneration processes also involve standardization, performance optimization, commercial scalability, innovative approaches to accessing encapsulated cells, and the incorporation of circular economy principles into battery design. Addressing these challenges is essential for realizing the full potential of battery regeneration as a sustainable and economically viable solution for managing end-of-life batteries.

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