Perspective

Prospects for managing end-of-life lithium-ion batteries: Present and future

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Abstract
The accelerating electrification has sparked an explosion in lithium-ion batteries (LIBs) consumption. As the lifespan declines, the substantial LIBs will flow into the recycling market and promise to spawn a giant recycling system. Nonetheless, since the lack of unified guiding standard and nontraceability, the recycling of end-of-life LIBs has fallen into the dilemma of low recycling rate, poor recycling efficiency, and insignificant benefits. Herein, tapping into summarizing and analyzing the current status and challenges of recycling LIBs, this outlook provides insights for the future course of full lifecycle management of LIBs, proposing gradient utilization and recycling-target predesign strategy. Further, we acknowledge some recommendations for recycling waste LIBs and anticipate a collaborative effort to advance sustainable and reliable recycling routes.

Key words
gradient utilization, lithium-ion batteries, predesign, recycling

1 INTRODUCTION

The advent of the electrification era has changed the global pattern of fossil fuel as dominating energy source. Electric vehicles (EVs) are seeing a surge in production and sales given the policy support, price cuts, and leverage in manufacturing technology.[1,2] BYD has officially announced that it will cease the production of fuel vehicles from March 2022, a feat that may inspire more automakers to follow suit. According to the forecasts by industry analysts, there will be at least 145 million EVs that will hit the road by 2030.[3] In addition to this, electrification has also penetrated ever deeper into various fields, such as energy storage base stations and portable devices, both of which carry massive lithium-ion batteries (LIBs). Strong momentum in electricity markets spurs the great demand for LIBs as well as the upstream raw materials. It has been estimated that the future production capacity of LIBs will need to reach 40 GW year\(^{-1}\) to meet the demand.[4] However, the calendar life of LIBs is generally around 8–10 years, and it is easy to foresee that there will be a steady stream of

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ex-service LIBs by then. Concerns have gradually surfaced in this context over how to usher and tackle the waves of used LIBs decently while maximizing the scale benefit and economic value.\[5\]

Concerning LIBs that have reached their limit of service life, land-filling or stockpiling is an inadvisable route.\[6\] For one thing, these two ways conceal a huge security risk, due to the nature of the fire-prone (or worse, explosive) LIBs. For another, inappropriate handling also leads to wasted energy and nonrenewable resources while also posing an environmental hazard, all of which run counter to the sustainable conception. Driven by maximizing utilization and cost-effectiveness, reliable and sustainable recycling emerges as the optimal solution for the rational disposal of massive end-of-life LIBs.

Owing to the confusion of battery lifecycle management and the absence of consolidated production criteria, the recycling market of waste LIBs is not yet mature. Herein, based on the concept of green and sustainable recycling, we expounded the orderly and hierarchical management strategy of decommissioned LIBs from refurbishment to reuse and finally to recycling, also the industry demonstration. Subsequently, the intelligent recycling-target predesign strategy is also elaborated to shape advanced LIBs with traceability and information visualization. Finally, we present some ideas on current recycling difficulties, which we believe will contribute to the sustainable recycling of spent LIBs over the coming decades.

### 1.1 Necessity of recycling spent LIBs

#### 1.1.1 Resource advantages

The geographical distribution of critical elements, particularly lithium and cobalt, is uneven, as shown in Figure 1A. Nearly 60% of the cobalt resources is located in the Democratic Republic of Congo, with Russia and Australia, which are tied for second place in terms of cobalt abundance, accounting for only 5% each. Lithium resources are 44% owned by Argentina and 34% by Chile. The distribution of nickel resources is slightly better. Moreover, continuous extraction aggravates the exploitation and depletion of the natural reserve.\[7\] In particular, mining and trading inevitably relate to sensitive political and human rights issues, resulting in the inability to...
guarantee a reliable supply of raw materials. Since enormous value is embedded in fabricated LIBs, feedstock sourced from spent LIBs fends off the reliance on imported upstream ingredients and realizes resource enrichment and secondary supply, which enhances supply chain resilience. Figure 1B shows that under the sustainable development scenario, the weight of lithium, cobalt, and nickel from the recycling pathway is projected to be 2, 8.2, and 31.1kt, respectively, by 2030, with an additional 0.8, 0.3, and 4kt of lithium, cobalt, and nickel expected to be generated through the reuse pathway, and more by 2040. This forecast is a strong indication that the recycling and reuse pathway for LIBs will significantly reduce the supply–demand for minerals and ease the tight supply chains, and that this contribution will continue to grow over time.

1.1.2 | Environmental protection

First and foremost is the enormous environmental impact of mineral extraction and smelting processes, such as soil erosion and dust phenomena. Second, there are hidden dangers of combustion or explosion in waste LIBs, and improper disposal methods will release a large number of toxic gases (like HF), liquids (organic electrolyte), and heavy metals (Co, Ni, etc.) into the environment, presenting serious air, water, and soil pollution that escalates environmental contamination. Proper recycling of waste LIBs contributes to the prevention of environmental pollution and the realization of the “dual carbon” goal.

1.1.3 | Economic attraction

The price of the strategic element cobalt is affected by several factors, such as politics, geography, and supply stability, and is highly volatile. And it is always at a high level by contrast with nickel (Figure 1C). However, the sudden price hike (up 250%) of nickel in early March this year also caused panic on the downstream demand side. On top of this, the price of Li$_2$CO$_3$ has also risen promptly since May 2021, up 150% year-on-year, with an unpredictable price spike (Figure 1D). Rising demand for lithium also appears to have exacerbated this trend (inset of Figure 1D). In this case, if the recovered metal materials were to re-enter the market economy, costs from mining, smelting, equipment maintenance, transportation, labor, and so forth would also plummet. Filling the resource gap could point directly to lucrative profits, which would be a profitable boon for both manufacturers and recyclers.

1.1.4 | Policy enforcement

Governments have been stepping up efforts to inch detailed regulations in response to the resource crisis and energy shortages. The European Union (EU) has drafted regulations that clearly emphasize the responsibilities and obligations of manufacturers, end-users, and recyclers, and has been striving to ensure transparency and richness of information throughout the lifecycle of batteries. The Japanese government attaches great importance to the recycling of used LIBs and has issued a number of relevant norms, with the nonprofit organization Japan Portable Battery Recycling Center (JBRC) specializing in this area. The United Kingdom has not yet implemented any regulations on the recycling of used LIBs, but it has proposed a tax on the use of batteries. The United States has enacted two federal laws related to recycling LIBs, Mercury-Containing and Rechargeable Battery Management Art and Resource Conservation and Recovery Art, both of which encourage the reuse of batteries. In early July 2021, China’s National Development and Reform Commission and others jointly issued a notice proposing to improve the traceability management system for the recycling and utilization of power batteries for EVs. Following that, the “Administrative Measures for the Echelon Utilization of New Energy Vehicle Theorem Batteries,” released in August, clarified the need to accelerate the construction of a power battery recycling system.

2 | CURRENT STATUS OF RECYCLING LIBs

At this stage, waste LIBs suffer from high disposal costs, low recovery rates, poor recycling efficiency, and inferior recycling profits. The background of the floundering recycling market in general lies in the chaotic internal structure and composition, poor lifecycle management, and untraceable battery information. While previous success stories of lead-acid batteries have boosted public morale for LIBs, LIBs face a much tougher recycling environment. LIBs are in a disordered state from design and application to recycling, there are no consolidated standards to navigate LIBs from cradle to grave, and the matching supervision is lax. It is imperative to blaze an exclusive and feasible recycling trail for LIBs, accompanied by maximizing economies of scale.

2.1 | Removal of the battery pack

Individual cells are commonly assembled into battery modules before being integrated into battery packs.
A battery pack is an energy storage device that includes battery modules, battery electronics, high-voltage circuitry, overcurrent protection devices, battery boxes, and interfaces with other external systems (e.g., cooling, high-voltage, auxiliary low voltage, and communications). Opening a battery pack is not an easy task. First, the exterior of the battery pack may be badly deformed or have been burned due to severe crushing or impact (e.g., in a car accident). Second, the bolts or other joints may also be rusty and difficult to remove due to aging. Not to mention wire welding and mechanical connections, common connection techniques used in LIBs, present great resistance to the removal of the battery housing. But the most difficult part is that battery packs come in all shapes and sizes, types, and weights, and the accessories used are not uniform. Disassembly requires specific unboxing tools, and efforts to overcome intractable soldering, intricate wiring, sealants (e.g., epoxy resin), and glues that securely hold individual cells in place. Exemplified by Tesla and Nissan, the former is intractable given the impregnable polyurethane cement holding the batteries tightly in place, while the latter takes around two hours to remove the rectangular Leaf battery module. These difficulties originate from the fact that only the reliability of product quality is taken into account during the manufacture of the battery packs, without considering procedures to facilitate battery recycling. Furthermore, vacancies in production standards and the unknown origin of the batteries led to an escalation in the difficulty of dismantling them. In addition to the enormous operational challenges, the potential fire hazards and toxic gas leakage hazards during dismantling pose a threat to the safety of the operators. Besides, at the current level, workers need to carefully and thoroughly survey the surface damage and internal energy of each unit to specialize in the treatment of the obtained batteries. And the possible safety incidents should be alert to at all times. Mechanically automated dismantling of battery packs is not yet commonplace, and the manual handling model creates a complex, cautious, time-consuming, and labor-intensive status quo.

Recently, there has been a wave of demodularization of battery pack assembly processes, with BYD’s “blade batteries” being the most prominent representative. The cells are arranged in arrays and embedded in the battery pack in the same way that “blades” are inserted into sheaths. Compared with BYD’s previous LiFePO4 batteries, the focus of the “blade batteries” upgrade is that the cells can be integrated directly into the battery pack (namely, cell-to-pack and CTP technology) without the need for modules, thereby greatly improving integration efficiency and avoiding tangles of wires and glue. Globally, Tesla, CATL, BYD, and SVOLT have announced and developed mass CTP-related production. By eliminating the battery module assembly process, the number of battery pack components is reduced by 40%, the volume utilization rate of CTP battery packs is increased by 15%–20%, and production efficiency is increased by 50%, significantly cutting the cost of manufacturing power batteries. The popularity of module-free power packs is on its way.

### 2.2 Acquisition of key materials

After cracking the stubborn battery pack, the central question arises as to how to access the precious raw materials contained in the battery. As cathode materials occupy a relatively high cost (about 30%–40%) of the entire battery, recycling is particularly favored with regard to them. The three main recycling levers available include the pyrometallurgical process, hydrometallurgical process, and direct recycling method (Figure 2), each with its own strengths and weaknesses.

#### 2.2.1 Pyrometallurgical process

The pulverized LIBs are melted at high temperatures in special furnaces, and eventually, metals such as Cu, Fe, Co, and Ni are recovered in the form of alloys, while lithium and aluminum remain in the slag. The incineration of anode materials, binders, electrolytes, conductive carbons, and so forth at high temperatures absorbs heat, which can be used to fuel the combustion process. The aluminum current collectors can act as fluxes, lowering energy consumption even more. Due to the low selectivity and sensitivity of the pyrometallurgical process to batteries (capable of handling mixed battery flows), and the fact that it does not require much human intervention, it can be widely used for all types of battery recycling and has been successfully implemented in commercial practice. However, this comes at the cost of energy intensity, harmful gas emissions, and more wasted resources. In addition, the resulting alloy products need to undergo further aqueous chemistry treatment and are incapable of recovering other low-value materials other than cathodes and Cu current collector, such as electrolytes, anodes, plastics, and so forth.

Most improvements in the pyrometallurgical approach have been aimed at lowering energy consumption, selective recovery, and improving lithium recovery. Zhang et al. treated waste LIBs by reduction roasting, where the cathode materials were dissociated and converted to elemental materials. Lithium could be preferentially extracted from the
roasted product by carbonated water leaching. Wang et al.\cite{23} demonstrate that the calcination temperature of LiCoO$_2$ could be decreased to below 400°C with the aid of (NH$_4$)$_2$SO$_4$. In the subsequent treatment, only water is used to dissolve the low-temperature roasting product, which further reduced the recovery cost. Lin et al.\cite{24} successfully achieved the selective recovery of lithium from LiNi$_{0.5}$Co$_{0.2}$Mn$_{0.3}$O$_2$ materials via a modified sulfate-roasting method. The yielded Ni$_{0.5}$Co$_{0.2}$Mn$_{0.3}$O$_{1.4}$ and Li$_2$SO$_4$ can be subsequently separated with water only. Pyrometallurgical recycling is well ahead in terms of its ability to handle mixed battery streams, but its recovery efficiency for critical metals is far from impressive and is usually supplemented by a hydrometallurgical process. In response, Stinn and Allanore\cite{25} demonstrated that several simple process levers (gas partial pressure, gas flow rate, and carbon addition) can selectively sulfide target metals from mixed metal oxide feed. Then they have sought to demonstrate the feasibility of 15 elements, including key elements contained in LIBs. Hence, a one-step selective recovery of transition metals is also a capital-saving option for pyrometallurgical recycling of LIBs.

2.2.2 | Hydrometallurgical process

Hydrometallurgy is another lever that dominates the recycling market and is widely applied in the Asian region in particular.\cite{26-29} After mechanical pretreatment, a combination of acids, bases, and reducing agents is usually adopted to dissolve the cathode materials, followed by subsequent purification and separation of the metals through precipitation, solvent extraction, and electrochemical routes.\cite{30-32} The hydrometallurgical process can maintain the high purity of the obtained material, and the low temperature operating also highlights the preponderance of safe recovery. Compared with the pyrometallurgical process, energy consumption and gas emissions have dropped a lot. Despite the energy and labor savings, the initial fragmentation renders the subsequent extraction of target elements tough. In particular, valuable metals, such as Co and Ni, strikingly resemble in nature, which upgrades the difficulty of the subsequent separation process. Besides, a large number of strong acids and bases used in this process imply an immense chemical input. This approach has certain environmental and economic limitations due to the substantial wastewater generated, as additional water pollution needs to be addressed.

Recently, in addition to using greener and degradable organics as leaching and reducing agents, researchers have sought to leverage more cost-effective and environmentally friendly leaching reagents such as deep eutectic solvents (DESS).\cite{33,34} DESS refer to the eutectic mixture of a proportion of hydrogen-bonded acceptors (such as quaternary ammonium salts) and hydrogen-bonded donors (such as amides, carboxylic acids, and polyols) with a freezing point significantly lower than the melting point of the individual.\cite{35} DESSs offer more prominent advantages over ionic liquid (IL), such as ease of preparation and low cost, and are often previously known for their powerful ability to dissolve oxides.\cite{36} Tran et al.\cite{37} used the DESSs consisting of a combination of choline chloride and ethylene glycol to initially dissolve waste LiCoO$_2$ materials with leaching efficiencies of >90% for both lithium and cobalt, which broadened the application of DESSs in the recovery of...
LIBs. Further, Wang et al.\cite{38} screened DESs composed of choline chloride and urea to realize the dissolution of LiCoO$_2$ at lower temperatures through Fukui function calculations and cyclic voltammetry measures. By adjusting the coordination environment of transition metals using different diluents, Chang et al.\cite{39} successfully achieved the selective separation of LiNi$_{x}$Co$_{y}$Mn$_{z}$O$_2$ (NCM, $x+y+z=1$) series of materials using DESs composed of choline chloride and oxalic acid. The combination of choline chloride and oxalic acid provides a good idea for the separation of cobalt and nickel, given the problems of their separation since ancient times. In addition, Kim et al.\cite{30} found that the deposition potentials of cobalt and nickel in concentrated chlorine electrolytes are totally discrepant from the ordinary dilute electrolyte. Supplemented by the electrode interface design, the efficient separation of cobalt and nickel is easily accomplished by electrolysis, which is also caused by the disparate coordination environments.

### 2.2.3 The direct recycling process

Direct recycling is dedicated to repairing the lithium loss and structural defects of active materials so that the cathode materials can be directly regenerated into a new electrode without being decomposed into elemental form. The nondestructive method is identified as an environmentally beneficial route due to its minimal energy and chemical input. After the discharging, the spent LIBs are manually sorted and disassembled, which can separate the intact electrodes, separators, and plastic casings. Its overall recovery rate is well received due to the availability of components other than the cathode, even the electrolyte. The damaged structure is rejuvenated by lithium replenishment, which is usually followed by a short annealing process to assist in improving crystallinity. The direct recycling method owns the maximal resource utilization and minimal environmental footprint but involves plenty of manpower in the intricate pretreatment process, which is why it is currently only in the laboratory demonstration phase, that is, it is labor-intensive and economically infeasible.

One type of tricky problem in the direct recycling process is the separation of the active materials from aluminum current collectors. Poly(1,1-fluoroethylene) (PVDF), a binder commonly used in cathode materials, is notoriously difficult to work with due to its particularly stable structure and properties. In contrast, detaching the graphite anode from the copper current collector is much easier because of the use of easily navigable binder blends, such as sodium carboxymethyl cellulose and polymerized styrene-butadiene rubber. For the time being, common methods of liberating the cathode active materials from aluminum current collectors include: (1) Burning off at high heat. PVDF starts to decompose at 350°C and completely at 600°C.\cite{40} During this process, the conductive carbon also disappears at high temperatures. Simple and swift, but energy-intensive. (2) Reagent dissolution. N, N-dimethyl formamide (DMF) and N-methyl pyrrolidone (NMP) have proven to be formidable foes for dissolving recalcitrant PVDF, but they are toxic and expensive.\cite{41,42} Bai et al.\cite{43} screened the green and safe triethyl phosphate (TEP) according to the Hansen solubility parameters to replace the highly toxic traditional solvents NMP and DMF. The cathode material was successfully liberated from the aluminum current collector without affecting the crystal structure and electrochemical performance. (3) Ultrasonication. Ultrasonication can assist to release the active substances from the current collector, but it is more suitable for laminated flat electrodes rather than roll-packed electrodes, and the separation efficiency needs to be raised.\cite{44,45} Chen et al.\cite{46} used a combination of ultrasonication and Fenton reagent to render PVDF viscous failure. A mechanistic study showed that the OH generated by the Fenton reagent under ultrasonic enhancement was effective in degrading the PVDF binder. (4) Cryogenic grinding. Wang et al.\cite{47} went the other way and implemented the pretreatment method of cryogenic grinding by exploiting the glass transition temperature of PVDF. The low temperature caused the organic PVDF binder to change from a highly elastic state to a glassy state, which compromised the performance of the binder. Furthermore, the glass-like binder breaks down due to its brittleness when subjected to external forces, so as to realize separation from the aluminum foil.

The direct recycling method aims to form a closed-loop recycling path by repairing the failures of LIBs, including lithium loss, phase transition, and the appearance of microcracks. The most common relithiation strategy is to mix the failed cathode material with a lithium source and then sinter it at a high temperature. Innovative relithiation methods have been coming up all the time. Shi et al.\cite{48} used the hydrothermal method to treat degraded NCM cathode material. The hydrothermal lithiation facilitated the successful recovery of the cathode to a high purity layered phase with a low cation mixing, thereby compensating for the compositional and structural defects of the ternary material. Subsequently, they took advantage of the properties of LiNO$_3$ and LiOH mixture to form molten salts at a lower temperature of 176°C to realize the relithiation of ternary materials under ambient pressure. This method also successfully demonstrated that the incorporation of lithium enables...
the transformation of the cathode material from the rock salt phase to the layered phase, and discards the high-pressure environment of the hydrothermal method, with potential for large-scale applications.[49] Using ILs with a wide liquid phase range and high thermal stability as solvents, Wang et al. [50] explored the effect of three different ILs on the direct regeneration of materials under ambient pressure via an ionothermal strategy, which provided new ideas for the relithiation of spent cathode materials. The direct regeneration method seems to be more suitable for LiFePO4 and LiMn2O4, as they relatively have low values and undergo only a single-phase transformation. But when it comes to complex and expensive relithiation environments, their cost advantage fades. Xu et al. [51] proposed a targeted healing method for failed LiFePO4 products, adopting a combination of LiOH and citric acid solution to restore LiFePO4 to health at low temperature and low pressure. The recovered LiFePO4 exhibited remarkable electrochemical performance even without annealing treatment. The method is safe, cost-effective, and highly workable. In addition to considering discarded cathodes as targets for lithium replenishment, researchers have also turned to graphite and separators. Using pre-lithiated graphite anodes or functionalized separators to directly match the degraded cathode to assemble a new battery not only replenishes the loss of lithium in the cathode but also alleviates the tedious preparation of new electrodes.[52,53] Moreover, by combining the characteristics of LIBs and dual-ion batteries, Meng and colleagues [54,55] prepared composite electrode from the used LiFePO4 cathode and used graphite anode. The prepared LiFePO4/graphite composite cathode material could realize a staged de-/intercalation reaction of Li+ and PF6− within a broadened electrochemical window. This idea not only unlocks the role of anions in the electrolyte and reduces the presence of inert components in the battery but also combines the high capacity of LiFePO4 with the high conductivity of graphite to achieve a higher energy density.

To meet the evolving market demands, Qian et al.[56] ingeniously converted the recycled ternary material into modern mainstream single-crystal high-nickel materials through a one-step molten salt method, effectively upcycling waste ternary materials. In this way, the new electrodes refurbished by the direct regeneration method were no longer constrained by the type of raw materials, and the single-crystal morphology repaired the micro-cracks that caused the polycrystalline cathode to fail. Coincidentally, during the same period, a similar approach was adopted by Ma et al. [57] to upgrade the waste Ni-lean ternary cathode toward Ni-rich single-crystal cathode materials. And further, given the poor traceability of the waste streams, they also successfully demonstrated the feasibility of this upcycling lever in dealing with mixed battery flows. On the other hand, there are also direct regeneration methods for converting waste cathodes into practical anode materials or high-voltage cathode materials.[58–60]

3 | DESIGN AND RECYCLING CONCEPTS BASED ON BATTERY LIFECYCLE

3.1 | Disposal of existing decommissioned LIBs

The scale of retired power batteries will surge after 2030.[61] For batteries that are not completely exhausted, it is a waste of resources and energy to directly judge them to recycle, and the best way is to continue their value. Dutch politician Ad Lansink is the first to put forward the waste management hierarchy in 1979, ranking waste management options from the most environmentally friendly to the least ideal.[6] Harper et al.[8] further extended this hierarchy into the field of battery recycling technologies, as shown in Figure 3A. Herein, we follow in the footsteps of giants and endow more meaning to the hierarchical management of the battery recycling stage.

3.1.1 | Refurbishment

When a battery pack is released, it must ensure a high degree of consistency in terms of capacity, internal resistance, voltage, discharge curve, and lifetime. Of course, there will always be some variation in performance between individual cells from the same batch. Initial inconsistencies accumulate with successive charge and discharge cycles, resulting in widening differences in individual cell parameters (state of charge (SOC), voltage, internal resistance, etc.). Similar to the shortboard effect in bucket theory, the worst-performing individual cell will determine the performance level of the entire battery pack, and prematurely damaged individual cells will certainly exacerbate the degradation of the pack in some cases. Thus, replacing prematurely failing cells will escort the subsequent gradient utilization. But battery parameters are not as visible as a plank of wood, so how do we pinpoint that short piece of wood? As the power battery itself is a complex electrochemical system, its capacity decay mechanism is affected by many factors, such as battery material, internal structure, self-discharge, and external environment, resulting in
differences in the degree of aging of individual cells, thus making it more difficult to accurately predict the remaining life of the battery.\[62\] Currently, the main research direction based on residual life prediction technology is battery whole lifecycle monitoring, that is, establishing a big data-based platform and approach for residual value analysis of retired batteries (such as integrating various methods based on the Kalman filter (KF) algorithm and its derivative algorithms) to realize real-time monitoring of SOC and state of health (SOH) of the battery.\[63\]-\[66\] By swapping out bad cells and reformulating the best match to bring the voltage, capacity, and internal resistance of the entire battery pack into alignment, the remaining energy can be used more efficiently. ReJoule’s technology\[67,68\] for diagnosing battery packs relies on electrochemical impedance spectroscopy (EIS), which uses alternating current swept at many frequencies to measure the health of materials within the battery. More advanced diagnostic techniques are expected to be developed in the coming years to provide data that can be interrogated when a battery pack is decommissioned to facilitate recombination or recycling.\[69\] Spiers New Technologies (SNT) characterized and classifies discarded automotive battery packs through a comprehensive diagnostic assessment, which can be refurbished for their original use or reuse for redeployment to a second life in other applications.\[7]\}

3.1.2 Reuse

When the capacity of the power battery decays below 80%, it is deemed unsuitable to support cruising. However, decommissioning is not the same as end-of-life. After retired power batteries have passed the residual energy test, they can still be used in different scenarios, such as energy storage, distributed photovoltaic power generation, household electricity, and low-speed EVs (Figure 3B), also known as the gradient utilization.\[70,71\] As a consequence, their unspent energy can continue to be tapped. The intermediate step of reuse prolongs their service life and delays the recycling deadlines, both as a positive response to calls for energy and resource conservation and to ease the pressure on the growing recycling market. B2U has begun in California trying to convert the battery packs that once powered Nissan Leafs to store energy for solar panels. The company says its solar panels can generate up to 1.65 megawatts of energy, while its retired Nissan Leaf batteries can retain 10 megawatt hours of storage—roughly the equivalent of a year’s usage in an American home.\[67\] In 2017, British company Powervault built a home photovoltaic energy storage system using retired power batteries from the Renault Zoe and Nissan Leaf. This system stores redundant photovoltaics generated by solar panels in the owner’s home and connects it to the
municipal grid. It can be called nearby during peak power consumption, reducing the cost of new substations and transmission equipment, and improving the flexibility of the local power grid. This energy storage system is currently being used in many homes and schools in the UK, where it can save homeowners up to 35% on their electricity bills. In Japan, ex-service LIBs that cannot afford to power EVs can still work for lighting, refrigerators in 7-11 convenience stores, mobile elevators as well as low-speed EVs, such as electric wheelchairs and golf carts.\(^{67}\) The potential for the reuse of retired LIBs for continuous energy supply is endless. As depicted in Figure 3C, The annual supply of secondary LIBs is large and expected to exceed 200 GWh by 2030. In this way, the additional energy provided by reuse drives the significant value of unlocking storage against the backdrop of growing demand for LIBs, alleviating pressure on electricity supply and reducing resource and energy waste. On the basis of its significant energy output and economic benefits, the prospects for reusing retired LIBs are very favorable.

In brief, power batteries in gradients utilization have a wide range of potential applications. It will also spread to provide energy for mobile charging piles and smooth out power fluctuations from distributed power sources, allowing for more efficient use of surplus energy.\(^{61}\) On the other hand, the reuse route is predicted to be effective in alleviating supply chain tensions (Figure 1B,C). Nevertheless, the untraceability of currently available waste streams, and the ambiguity of the relevant information, make it difficult for recyclers to accurately evaluate SOH and therefore judge whether the obtained LIBs are worthy of secondary employment.\(^{72}\) The underlying cause of this difficulty is the lack of uniform criterion to guide the full lifecycle of LIBs, resulting in the varying internal structures and chemical compositions of LIBs circulating in the marketplace. In addition, it remains controversial as to who guarantees the quality of LIBs for secondary use.

The ultimate destination of end-of-life batteries is certainly and inevitably recycling. As the LIBs recycling segment continues to heat up, domestic and international recycling companies that used to provide only battery raw materials are actively extending downstream in the industrial chain. Li-Cycle, the largest recycler of LIBs in North America, puts end-of-life LIBs into a container that can be discharged and shredded simultaneously, recovering different metals through a staged hydrometallurgical process that will facilitate the large-scale dismantling of LIBs in years to come.\(^{73}\) Redwood Materials combines pyrometallurgical and hydrometallurgical methods. First, the residual power of the waste LIBs is used to drive the converter to generate a high temperature, then the metals are separated by the pyrometallurgical method. Finally, various metal feedstocks are recovered and purified by the hydrometallurgical method.\(^{74}\) SungEel Hitech, the largest battery recycler in Korea, currently can process 24 000 tons of LIBs materials per year and plans to increase its annual production to 56 000 tons through the hydrometallurgical process to recover nickel, cobalt, manganese, lithium, and other metal materials. The company has entered into a partnership with EcoGraf, an Australian graphite material company, to drive the expansion of the recycling business into high-purity graphite materials by the end of 2020.\(^{75}\) As a global material technology group, Umicore of Belgium has made recycling one of its four core businesses. Since 2018, Umicore has successively reached cooperation with BMW, Audi, and Swedish battery company Northvolt in battery recycling. Umicore is responsible for recycling valuable metals from LIBs and reprocessing the recycled materials into precursor and cathode materials.\(^{76,77}\) In China, GEM is currently the world’s largest manufacturer of LIBs recycling and one of the world’s largest sites for cobalt-metal refining and nickel-metal recycling. GEM plans to have an annual recycling capacity of 350 000 sets of power batteries after 2023.\(^{78}\) According to publicly available information, Brunp Recycling recycles and treats more than 6 000 tons of used batteries annually.\(^{79}\) In addition, companies such as Ganzhou Highpower, Guangdong Guanghua Technology, Quzhou Huayou Cobalt New Materials Co., Ltd., and Changqing New Energy continue to join in the recycling field, which will also promote the healthy and sustainable development of China’s new energy industry. However, based on the imperfection of the current recovery system, the overall recycling rate is still at a relatively low level, as illustrated in Figure 3D. The recovery rate for cobalt is only 32%, while the recovery rate for lithium is a pitiful 0.5%, and the recovery levels of other essential elements for LIBs, such as nickel, copper, and aluminum, are not satisfactory either. Therefore, it is necessary to vigorously develop advanced technologies to improve the recovery rate and recovery efficiency to meet the overall demands of sustainable development and a circular economy.

So far, most research on waste LIBs has focused on the recovery of Ni, Co, and Li elements that fetch a high price. In contrast, relatively little attention has been paid to lower value-added components, such as anodes and electrolytes.\(^{80}\) The recovered graphite anodes vary in terms of purity and structural integrity due to different recovery methods and processes. They can be used as raw materials with low requirements for electrochemical performance, or they can be used to prepare value-added graphene.\(^{81-83}\) Electrolyte recovery techniques include
organic solvent extraction, vacuum pyrolysis, and carbon dioxide supercritical extraction.\textsuperscript{184-188} More recently, Zhao et al.\textsuperscript{189} took a convenient approach to dismantle spent LIBs straightforwardly in water by abandoning the cumbersome process. The simultaneous collection and precise separation of the anode and the electrolyte can be realized without undergoing discharge.

\section*{3.2 Recycling-oriented predesign route}

The disposal of spent LIBs may seem to be a terminal issue, but it actually serves as a warning of the original intention for which LIBs are currently designed. The reuse and recycling of LIBs encounter a high technical threshold due to their complex composition.\textsuperscript{90} Advanced technologies, such as digital twin systems, artificial intelligence (AI), the Internet of Things (IoT), and machine learning (ML) are flourishing in the engineering field.\textsuperscript{117} Ideally, the promotion of high-tech concepts in the battery sector promises high transparency, information sharing, and traceability, sustaining the development of LIBs.\textsuperscript{4}

In realistic scenarios, various off-line evaluation measures, such as Coulomb counting, open-circuit voltage (OCV), and EIS are used to determine the SOC, SOH, and remaining useful life (RUL) of the batteries.\textsuperscript{40,91} However, these evaluation methods lack accuracy and precision given the real-world variation in the various battery aging mechanisms, ranging from inconsistencies in production and manufacture, and vastly different vehicle operating conditions, to small changes in temperature, solid electrolyte interface (SEI) growth, lithium deposition, active material dissolution, and electrolyte decomposition.\textsuperscript{92} A digital twin system is a cloud-based digital virtual model created from real-world physical entities. The model can map and simulate entities in real-time from multiple dimensions and has a wide range of applications in battery state estimation, fault diagnosis, and cloud control.\textsuperscript{93,94} The digital twin can reproduce the inside of a battery pack very accurately. By embedding IoT sensors with radio frequency identification (RFID), battery health can be monitored in real-time.\textsuperscript{95,96} The built-in sensor can decode chemical and thermal events in the battery and collect the data covering all battery health-related information, such as charge and discharge curves, temperature evolution, internal resistance, SOC, and OCV over its entire lifecycle, which is then uploaded to the IoT and stored in a powerful cloud database.\textsuperscript{97}

As an automatic identification technology, RFID carries out noncontact, bidirectional data communication through radio frequencies, which are used to read and write recording media (electronic tags or radiofrequency cards) for the purpose of target identification and data exchange.\textsuperscript{98,99} By replacing the QR codes on modern batteries with RFID tags, manufacturers and recycling authorities will be able to record and monitor the health of batteries remotely.

The next stage is to use the powerful computing and storage capacities of the cloud to comprehensively collect the data over the lifecycle of the batteries. The digital twin system uses ML algorithms to compare and analyze historical and real-time battery data.\textsuperscript{92,100} From the digitized information collected, ML accurately analyzes the operational status of the battery pack, calculates control parameters that can be fed back to the battery management system, such as performance and safety monitoring, aging and health predictions and temperature evolution, and then provides early warning of potential hazards.\textsuperscript{97} Well-trained ML techniques, combined with powerful cloud computing and storage systems, are expected to facilitate highly accurate and cost-effective battery management systems.\textsuperscript{101}

In addition, incorporating sustainability and recyclability into the origin battery design will greatly facilitate the orderly management of batteries during in- and ex-service periods. Ideally, the decisive step in battery recycling would depend primarily on the ability to automatically sort and dismantle the batteries. By accessing the battery’s RFID tag, not only the recyclers but also the robots would be able to acquire information about the batteries, such as source, type, experience, and SOH.\textsuperscript{17,95} Precise instructions from the digital twin system enable the robots to sort, nondestructively disassemble and separate critical materials from end-of-life LIBs with detailed guidance.\textsuperscript{92} With this line, automatic mechanical dismantling enables recyclers to cope with countless waste flows, saving labor and time with considerable efficiency and profitability. Additionally, the enhanced level of automated dismantling also reduces the risk of human injury, thereby reducing labor costs, and the improved return on capital makes recycling more economically attractive.\textsuperscript{102,103} Currently, automated dismantling robots are still in their infancy. Automated robots are still highly dependent on a structured environment, that is, pre-programmed repetitive actions on precisely known objects at a fixed location.\textsuperscript{8} The deployment of intelligent automated disassembly not only poses a significant challenge to the mechanical automation discipline but furthermore provokes public thinking about battery design and calls for the introduction of uniform production standards, including battery chemistry, battery attachment and fixation methods, and the use of adhesives.

To summarize, the information provided by the smart battery design system makes it possible to track the flow
of LIBs (as demonstrated in Figure 4), allowing for targeted recycling efforts. The battery information is digitized in real time and uploaded to a shared cloud-based database through the ubiquitous IoT, and the health and operational status of the batteries are then monitored online through AI, and diagnosed and predicted. By reading the information about batteries that have been eliminated, the robots can judge, screen and sort the next flow of batteries, and follow the hierarchical management route and implement recycling under clear instructions. It is believed that in the near future, the intelligent management system can serve the entire lifecycle of new LIBs from manufacturing to assembly and make appropriate decisions on the fate of discarded LIBs, thus cutting the recycling costs, enhancing recovery efficiency, and accelerating the operation of the circular economy.

4 | LOOKING AHEAD

The first batch of power LIBs will enter a phaseout period as a result of the booming production of LIBs, and how to properly handle the eliminated LIBs has become a focus of attention. Recycling provides an additional opportunity for secondary supply and further lowers the manufacturing cost for LIBs, which is in line with the sustainable and reliable development strategy of resources and energy. Governments in a number of countries have also begun to value the recycling of LIBs, and have drafted or promulgated a series of regulations and policies, which provide a strong foundation for the orderly advancement of recycling.

Of the common recycling routes, the pyrometallurgical method owns immense energy consumption and greenhouse gas emission issues, but it has the ability to recover mixed battery streams. Improvements in the pyrometallurgical method are directed at boosting the recovery of lithium from slag, and focusing on the issue of greenhouse gas emissions. The drawback of hydrometallurgical recycling lies in the excessive chemical inputs and the subsequent troublesome effluent treatment. The search for green and cost-effective solvents appears to alleviate environmental pressures. The direct recycling method is more in line with the current concept of green sustainability and circular economy, but the labor costs involved are too high. In addition, direct recycling should also take into account the upcycling of feedstock, so that it is not bound to the limitations of raw materials and specific products and better comply with the signals sent by the market. Furthermore, recycled materials are generally considered to be inferior in performance to their commercial counterparts, and the data provided during laboratory demonstrations is deemed unreliable by the public. Researchers have had reservations about this. After treating waste ternary cathodes by hydrometallurgical method, Ma et al. readjusted the element ratios, prepared new cathode materials by co-precipitation, and tested their electrochemical performance under...
harsh industrial conditions. It was found that the unique void-rich structure endows new cathodes with better electrochemical performance. This experiment not only dispelled the doubts from the industry about the properties of the recycled materials but also builds a bridge for the dream linkage between industry and academia. In addition, more attention should be paid to simplifying battery assembly and the separation of internal components, with continued innovation and in-depth exploration of sustainable recycling approaches.

The rise of a large number of LIBs has also put considerable pressure on the recycling market, where the level of technology is not yet perfect. Therefore, it is advocated that primary LIBs enter the reuse staging area and get a second chance to serve and explore their residual value before entering the recycling phase. After diagnosis and refurbishment, the reborn LIBs will be allocated to energy supply scenarios, such as low-speed EVs, energy storage base stations, and valley power peaking, maximizing the utilization of energy and resources. The gradient utilization extends the service life of LIBs and relieves the pressure on recycling, but there is a controversial issue of who is responsible for the quality and regulation of secondary utilized LIBs?

All LIBs that reach the end of life will eventually flow into the recycling phase. We demonstrated a predesign route based on intelligent battery lifecycle management. With the implantation of high technology, the recycling-target predesign not only enables the traceability of the waste obtained but also enables comprehensive real-time monitoring of the battery status, making the battery information transparent and visual. In addition, with the assistance of automated disassembly, high efficient, and profitable recycling management is within reach. In summary, a management plan that runs through the entire lifecycle of the battery has been formed, as shown in Figure 5. That is, through the elaborate recycling-oriented pre-design, the intelligent technology will be integrated into the battery management system and continuously output battery-related information. After the LIBs have finished their first phase of service, the recycling organization will evaluate and refurbish the LIBs based on the valid information provided by the intelligent management system, and then reintroduce them to the reuse market. When the LIBs have run out of their last energy, automated robots will come out to dismantle the end-of-life LIBs automatically under the precise instructions of the intelligent system and reintegrate them into the circular economy in the form of new raw materials. It is believed that this vision is achievable in the near future through joint efforts. From the current point of view, the promotion of a sustainable recycling chain for end-of-life LIBs can also be approached from the following points of view:

1. Set relevant standards: The disruption in the LIBs market is largely due to the lack of standards to discipline and regulate the production, assembly, management, and recycling of LIBs, which is a pressing issue that needs to be addressed. Under the support of formalized standards and strengthened supervision, the LIBs market will be nurtured to an orderly pattern and strict discipline. Further, technologies, incentives, and subsidies need to be enacted to instrument sustainability in recycling.

2. Scaling-up: The primary factor driving the evergreen recycling segment for recyclers is sustainable and substantial profits. However, given the current tedious, expensive, and energy-intensive recycling process, the use of new, cheaper, and more reliable feedstocks eclipses the recycled materials. The key to yielding long-term economic returns is to expand the recycling scale to meet the high throughput of LIBs in the market, which can also offset the higher raw materials costs.

3. In situ disposal: Regardless of the economic aspect of reducing transportation costs or the safety aspect of reducing hidden dangers, “disposal on site” is a more appropriate option than transporting used LIBs to less stringent countries. It is highly desirable to popularize the local recycling stations and gradually build up the entire infrastructure around waste LIBs recycling. Passivation or dismantling of waste LIBs locally or through short-range transport will also increase the recovery rates, lower the transportation cost, and enhance the recycling efficiency of waste LIBs. Furthermore, in-situ disposal also makes it
possible to implement deposit refund measures for small LIBs and therefore increase the recycling rates.

4. Technological improvement: Nowadays, LIBs have seen a proliferation of innovations in materials and designs, for instance, the nascent high-nickel single-crystal electrode materials, the boom in silicon anodes, and the emergence of 4680-type batteries. However, the level of recycling LIBs has not kept pace. The mechanical what-comes-in-what-gets-out recycling model also tends to cause a disconnect with the evolving LIB market. This calls for improvements to current recycling methods, ensuring low energy consumption, low environmental hazards, and less manual intervention while increasing the ability to handle mixed waste streams and value-added recycling. On the other hand, achieving intelligent management and automated nondestructive dismantling of LIBs from production to end-of-life will require relentless efforts from all parties.

5. Strengthen cooperation between academia and industry as well as collaboration between enterprises: To circumvent the limitations of recycling technologies development caused by information mismatches between academia and industry, viable laboratory-level outcomes need to be brought to plant-scale for volume implementation. Second, the dynamic evolution of the composition and structure within the LIBs requires the enterprises to keep pace. Close collaboration between enterprises can facilitate information sharing and technology exchanges, further leading to shaping a robust business model for mutual benefit.

6. Improve and simplify the way LIBs are manufactured and assembled: The removal of binders has been an obstacle to obtaining nondestructive materials. The development of binders that are soluble in nontoxic and readily available green reagents (such as water or ethanol) will replace the stubborn PVDF binders without compromising battery performance.[105-108] In this way, both the cost of manufacturing and recycling electrodes will be reduced, and the manufacturing and recycling steps will be simplified, with a consequent increase in safety. On the other hand, streamlining the production and assembly procedures would also facilitate automated disassembly. Exemplified by the aforementioned CTP technology, demodularization renders the assembly and disassembly of the battery pack simple and fast.[3,7]

7. Develop other sustainable batteries: For the moment, the landscape of LIBs is dominated by the NCM series and LiFePO4, with slow uptake of all-solid-state batteries and aqueous LIBs.[109,110] The academia and enterprises have joined forces to prosper other secondary alkaline ion batteries, such as sodium-ion batteries (CATL, HiNa Battery, Sunwoda) and potassium-ion batteries.[111-115] The aim is to take the load off the overburdened LIBs in certain cases, while advancing green, safe, and diversified alternatives for energy supply, also as a driver for sustainable development.[116,117]

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CONFLICT OF INTEREST
The authors declare no conflict of interest.

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