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Sustainable value chain of retired lithium-ion batteries for electric vehicles

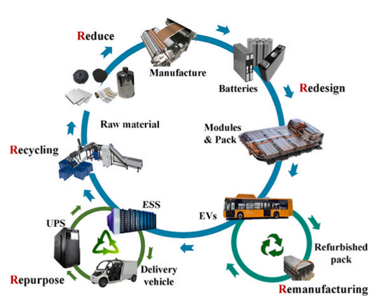
Yang Hua [a], Sida Zhou [a], Yi Huang [a], Xinhua Liu [a,b,**], Heping Ling [c], Xinan Zhou [a], Cheng Zhang [d], Shichun Yang [a,*]

[a] *School of Transportation Science and Engineering, Beihang University, Beijing, China*
[b] *Dyson School of Design Engineering, Imperial College London, London, UK*
[c] *Automotive Engineering & Research Institute, BYD Automobile Industry Co., Ltd., Shenzhen, 518118, China*
[d] *Institute for Future Transport and Cities, Coventry University, UK*

HIGHLIGHTS

- Systematic summary of the sustainable value chain of lithium-ion batteries.
- A 5R principle in the circular value chain process is demonstrated.
- Comprehensive analysis of reuse and recycling from different perspectives.
- Remaining challenges and perspectives of circular value chain are discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Lithium-ion batteries (LIBs) have been widely used in electric vehicles due to the advantages of high energy/power densities, high reliability and long service life. However, considering that a massive number of LIBs will likely retire and enter the waste stream in the near future, the handling of end-of-life LIBs must be taken carefully. The effective utilization of retired LIBs, which still remain about 70–80% of the initial capacity, can extend battery life, conserve natural resources and protect the environment. Herein, this review provides a systematic discussion on the circular value chain (CVC) of spent LIBs, and proposes a 5R principle entailing reduce, redesign, remanufacturing, repurpose and recycling in the CVC process. Then the state-of-the-art technologies for remanufacturing, and a thorough summary of key issues and applications of repurpose process, are presented in detail. Subsequently, this article presents a comprehensive discussion on the recycling process, including pre-treatments and mainstream recycling technologies, from the prospects of technical, economic and regulation perspectives. Advanced technologies such as big data, block chain and cloud-based services, as well as the improvement of regulation and standardization processes, are required to solve the issues. Finally, the future challenges and prospects for sustainable CVC are highlighted.

* Corresponding author.
** Corresponding author. School of Transportation Science and Engineering, Beihang University, Beijing, China.
E-mail addresses: liuxinhua19@buaa.edu.cn (X. Liu), yangshichun@buaa.edu.cn (S. Yang).

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1. Introduction

Due to the concern of energy problems and environmental sustainability, electric vehicles (EVs) are becoming increasingly popular in this decade. As the energy supply component in EVs, battery packs are typically consisting of hundreds or even thousands of cells connected in parallel and series. Lithium-ion batteries (LIBs) have been widely used as EV power systems due to their advantages of high energy/power densities, long service life, and low self-discharge rate [1]. Annual sales of EVs reached 2.2 million in 2019 [2], and it is estimated that the EV LIBs market will exceed $30 billion and reach dozens of GWhs annually by 2020 [3]. LIB packs are expected to serve in EVs for about 8–10 years, and then should be replaced when they reach 20% capacity loss [4,5]. The global annual weight of retired LIBs may exceed 500,000 tons in 2020, and the retired EV packs can reach 6.8 million units by 2035 [6]. Assuming the average weight of a pack is 250 kg [7], the weight of spent EV batteries worldwide can weigh 1,700,000 tons by 2035.

Retired batteries still remain 70–80% of the initial capacity and have the potential to be utilized in less-stressful demanding applications [4]. Furthermore, spent EV LIBs contain many valuable resources such as lithium (Li), cobalt (Co) and manganese (Mn) [8], which can be recycled to reduce the resources requirement, and the global business of retired LIBs is estimated to reach $3 billion by 2035 [6]. Therefore, the handling of end-of-life (EOL) LIBs must be taken carefully. The effective utilization of retired LIBs can extend battery life, conserve natural resources and protect the environment [5].

The traditional approaches of handling retired LIBs can be divided into disposal, recycling and reuse [4,7]. Disposal means that the retired LIBs will be abandoned and discarded, which will release pollutants to environment [9]. The heavy metals such as Co and nickel (Ni) may adversely affect environmental quality and human health. The lithium salts such as $LiPF_6$ and $LiBF_4$ are toxic and corrosive to eyes and skin. In addition, disposal means a great waste owing to the loss of valuable materials such as Co and Li. According to the U.S. geological survey, the world's terrestrial reserves of Co and Li are limited to approximately 7 million tons [10] and 62 million tons [11] respectively. Driven by the strong demand of EVs, the prices of Co and Li have risen sharply in recent years [12]. Battery recycling is of great significance for sustainable development. Recycling process can separate the retired batteries into different components and extract the precious materials into the value chain [8,13]. Valuable resources such as metals and cathode active materials can be recovered using physical and chemical methods [9]. Reuse, including remanufacturing and repurpose, means that the qualified retired LIBs can be used in different applications such as automotive service, energy storage system (ESS), photovoltaic (PV) energy, and residential services depending on the evaluation results [14,15]. Due to economic and environmental advantages, priority should be given to reuse and recycling processes rather than disposal. Because of the different structure, various composition, and different operating histories, the handling of retired batteries must face the technical, economic and environmental challenges [6,14].

In this review, the circular value chain (CVC) of retired EV LIBs are introduced. To maximize the economic, environmental and resource benefits of the sustainable CVC, a novel "5R" strategy is proposed, namely remanufacturing, repurpose, recycling, reduction and redesign. The state-of-art technologies for remanufacturing are discussed in detail. A summary of improvement and challenges of the repurpose process are presented from the technical, economic and market perspectives. Then we discuss the key issues in recycling process comprehensively, including pre-treatments and current recycling technologies. Finally, the future challenges and prospects for sustainable CVC are presented. With the help of advanced technologies such as big data and cloud computing, 5R principle can provide effective guidance for the large-scale industrial development of EV and energy storage applications in the CVC process.

2. Value chain of retired lithium-ion batteries

2.1. Structure and composition of EV battery packs

In EV applications, different automotive original equipment manufacturers (OEMs) have adopted various pack solutions with different physical configurations, module structures, battery shapes and internal chemistries.

An EV pack usually consists of multiple modules, which are composed of multiple cells. Shapes of EV LIBs can be divided into a cylindrical, prismatic and pouch geometry [16]. The cylindrical cell is a widely used packaging style for EV due to the advantages of low cost, ease of manufacture, and good mechanical stability. For instance, Tesla Model S uses thousands of cylindrical cells to form battery packs between 75 and 90 kWh [17]. However, the packing density and weight efficiency of cylindrical cells may be limited due to space cavities from the point of geometry, and large number of cells also increase the integration and assembly cost [18,19]. Therefore, OEMs are tending to use prismatic or pouch designs to optimize the system-level packaging efficiency. For instance, BMW i3 uses Samsung/SDI prismatic cells [7] and Nissan Leaf adopts pouch cells [20]. The prismatic cell can improve space utilization and therefore has the advantages of high weight and volume efficiency, flexible design and low assembly cost [21]. The disadvantages of prismatic cell include high production costs and high capacity fading rate caused by temperature gradient [22]. The pouch cell uses soft packaging and has the advantages such as high safety, low mass, high packaging efficiency, and improved heat dissipation performance [19]. However, the pouch cell suffers from weak mechanical strength, thus needs a special support design to avoid swelling [22].

LIBs are composed of anode, cathode, electrolyte, separator, current collector and container [12]. The anode is usually made of graphite or lithium-titanate oxide (LTO); lithium metal oxides are usually used as the cathode, such as lithium iron phosphate (LFP), lithium manganese oxide (LMO), lithium cobalt oxide (LCO), lithium-nickel manganese cobalt oxide ($LiNi_xMn_yCo_{1-x-y}O_2$, NMC) and lithium nickel cobalt aluminium oxide (NCA) [12]. The electrolyte is usually made of organic solvents (such as ethylene carbonate and diethyl carbonate) [23], lithium salts (such as $LiPF_6$ and $LiBF_4$) and additives [24,25]. The separator is a microporous membrane usually made of polypropylene (PP) or polyethylene (PE) et al., which prevents direct contact between the cathode and anode [26,27]. The current collectors are usually made of metal materials such as aluminium (Al) and copper (Cu) [28,29]; and Al is the primary material used for battery containers [30,31].

Although EV OEMs have adopted different solutions for battery pack design, the production processes of different LIB cells are very similar. Generally, the cell manufacturing process consists of multiple steps such as mixing, coating, calendaring, slitting, winding, welding and testing, etc. [32]. Then the LIB cells are connected in parallel and series to form modules and packs, which can be equipped in EVs as energy supply.

2.2. Circular value chain of retired lithium-ion batteries

EVs can travel 120,000 to 240,000 km throughout their whole lifespan [33], and the performance of EV LIBs degrades over time. Therefore, a large amount of EV LIBs will retire and enter the waste stream in the near future [34]. The CVC of retired LIBs can be seen in Fig. 1. The profitable treatment of retired EV LIBs includes remanufacturing, repurpose, and recycling, which are located at different positions in the CVC. To capture the maximum value of LIBs, the ideal solution is to remanufacture the retired EV LIBs, then repurpose the cells, and finally obtain valuable materials by the recycling process.

The CVC starts with battery evaluation, which determines the value of the retired EV LIBs and their suitable applications. The highest rated LIBs can be remanufactured in automotive scenarios for OEMs or spare parts market [36]. The medium-score LIBs can be repurposed in less demanding scenarios such as grid-related ESS, electric delivery vehicles,

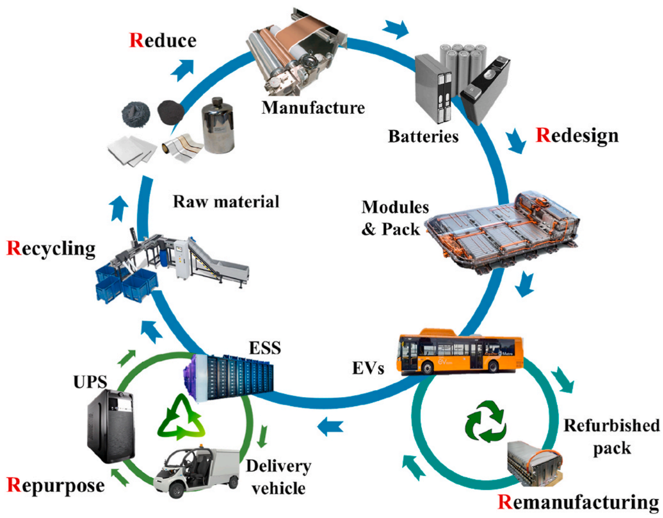

Fig. 1. Circular value chain of retired lithium-ion batteries [12,24,35].

and uninterruptible power system (UPS), depending on the evaluation score [37]. LIBs below the reuse standard will be recycled to obtain valuable materials. It should be noted that the remanufactured LIBs can be repurposed after further aging, and those repurposed batteries will eventually be recycled after further degradation, thereby achieving a closed-loop CVC. In addition, redesigning the current battery pack during the manufacturing process can help improve utilization rate and increase convenience, and technological advances can reduce the demand for raw materials and accessories [8]. Remanufacturing, repurpose and recycling, as well as reduction and redesign, can form a "5R" strategy for managing retired battery to maximize the economic, environmental and resource benefits.

Disposal is another possible treatment for retired EV LIBs. Though some of the disposal batteries are sent to waste-to-energy plants for incineration to generate energy, the majority of the disposal LIBs are sent to landfills directly [28]. Due to unnecessary material waste and environmental threats, disposal is the most undesirable option in EV LIBs processing [24], therefore not considered in the CVC process.

3. Remanufacturing

After losing about 20%–30% of the initial capacity, LIBs can hardly satisfy the power and energy requirements of EVs such as driving range and acceleration [38]. These retired LIBs are still attractive to remanufacture or repurpose in various applications due to the high remaining capacity [39,40].

The EOL batteries are firstly collected and transported to the processing plant, and then evaluated to determine the next treatment procedure. The battery evaluation depends on several factors, including the state of health (SOH), remaining useful life (RUL), safety level, as well as the specific cost and benefits [8]. The LIBs with the highest evaluation score can be remanufactured for EV applications. The schematic flowchart of the remanufacturing process is shown in Fig. 2.

The remanufactured LIBs must meet all the requirements and standards specified by the EV OEMs, such as available capacity, power, and RUL. Due to cell-to-cell variations, the LIB cells within the same pack usually degrade at different rates. The retirement of the pack is often caused by the failure of some special cells, which may account for only a small part of the battery pack [15,41]. With the help of screening technology, the degraded batteries can be identified and replaced with new or qualified cells from other packs. Therefore, a batch of EOL EV packs can be converted into a smaller number of qualified packs through remanufacturing [42]. As shown in Fig. 2, the remanufacturing process usually includes a comprehensive battery test (assessment), screening,

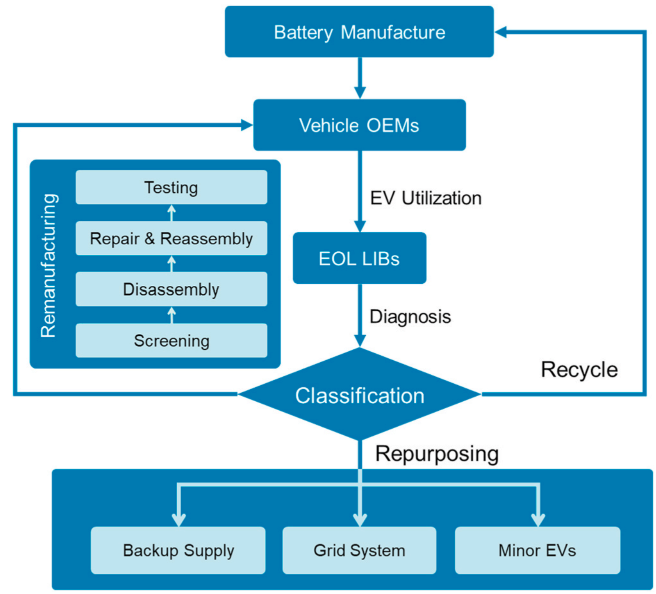

Fig. 2. Flowchart of the remanufacturing and repurposing process.

partial pack disassembly, removal and replacement of degraded cells (repair), and reassembly of the pack [43]. Then the remanufactured packs can be sent to the automotive OEMs or spare parts market after thorough testing [8,36].

3.1. Assessment

The assessment process can help evaluate the value of the EOL LIBs. Generally, safety level, SOH, and RUL are the most critical evaluation indicators for the spent EV LIBs [44,45].

3.1.1. Safety evaluation

LIBs abuse can be classified mechanical, electrical, environmental and chemical [46]. As shown in Table 1 [47], many organizations have

Table 1
Examples of LIB safety standards proposed by international organizations, adapted from [47].

Test	UL		IEC		SAE	IEEE	
	1642	2054	62,133	62,281	J2464	1625	1725
External short circuit	✓	✓	✓	✓	✓	✓	✓
Abnormal charge	✓	✓	✓	✓	✓	✓	✓
Forced discharge	✓	✓	✓	✓	✓	✓	✓
Crush	✓	✓	✓		✓	✓	✓
Impact	✓	✓		✓		✓	✓
Shock	✓	✓	✓	✓		✓	✓
Vibration	✓	✓	✓	✓	✓	✓	✓
Heating	✓	✓	✓		✓	✓	✓
Temperature cycling	✓	✓	✓	✓	✓	✓	✓
Low pressure (altitude)	✓		✓	✓	✓	✓	✓
Projectile	✓	✓				✓	✓
Drop				✓			
Continuous low rate charging				✓			
Penetration					✓		
Separator shutdown integrity					✓		
Internal short circuit test	✓						

established specific testing standards for EV LIBs to ensure the safety of drivers and passengers. These standards cover the majority of the safety risks such as external short, crush, penetration, heating and abnormal charge.

However, such abuse test will lead to irreversible damage to the EV LIBs, and some specific application issues may also escape the standards. Therefore, it is necessary to evaluate the pack safety performance based on non-destructive testing methods to support remanufacturing and repurposing.

Model-based and data-driven methods have been developed to evaluate the safety characteristics of EV LIBs. Wang et al. [48] proposed a soft short circuit failure model based on second-order oscillation feature to evaluate the safety performance and predict the failure possibility. The warning system can predict the soft short circuit failure with an error less than 3.5%. Li et al. [49] developed a detailed computational model of pouch LIB with high accuracy, and the safe operation range (safety envelope) of mechanical loading conditions can be obtained with machine learning algorithm, which can be used to predict the safety of energy systems.

Assessment techniques such as state of safety (SOS) estimation are also used for safety evaluation of aged EV batteries [50], and the relationship between SOS function and different abuse conditions can be seen in Fig. 3. However, the practicability of SOS estimation methods needs further verification.

3.1.2. SOH evaluation

SOH can quantify the degree of capacity decrease and internal resistance increase, which may be caused by complex side reactions [45]. The major side reactions [32] are shown in Fig. 4 [51].

The SOH estimation methods can be divided into the following categories, including direct assessment approaches, incremental capacity analysis (ICA) based methods, empirical methods, equivalent circuit models (ECMs), electrochemical models (EMs), data-driven methods and hybrid methods [45,52,53].

Wang et al. [54] experimentally characterized the LFP battery's capacity loss under different operating conditions, and derived a capacity degradation model based on a cycle-test matrix including temperature, depth of discharge (DOD) and current rate. Empirical methods are easy to calculate, but their accuracy is limited to used cell chemistry and the tested operating conditions. Yang et al. [55] studied the aging behaviour of LIBs based on an electrochemical-thermal-mechanical coupled model, and established the relationship between the rate of side reactions and working conditions. Experimental results showed that the proposed method provided an accurate SOH prediction. However, the high computational cost limits the online application of the EM-based models. Yang et al. [56] proposed a SOH estimation method based on a three-layer back propagation (BP) neural network (NN). The ECM parameters including state of charge (SOC), ohmic resistance, polarization resistance, and polarization capacity were used as the input layer. The NN model could estimate the SOH with low computation cost, and the training time of NN was about 21 ms. Static and dynamic current profile tests verified the high accuracy of the NN-based method, and most of the SOH estimation errors were less than 5%. Li et al. [57] combined the grey relation analysis (GRA) and entropy weight method (EWM) to assess SOH. The GRA method was used to extract health indexes based on ICA curves, and the EWM was applied to evaluate the weight of such health indexes. Then SOH was obtained by calculating the grey correlation between the comparative sequences. Experimental results showed that the SOH estimation error was lower than 4% and the average error was about 0.6%. Ladpli et al. [58] presented a SOH estimation method based on acoustic-ultrasonic guided waves. Preliminary experiments on pouch LIBs showed that the information such as time of flight and signal amplitude varied with the aging process, which was used for accurate SOH estimation through statistical analysis.

Although many SOH estimation methods have been developed, most of them have not been widely adopted in real applications. The accuracy of SOC-based or ICA-based methods is still unsatisfactory, and the data-driven methods may encounter robustness problems in practical applications [59]. Therefore, the hybrid methods combining different approaches may be a promising direction to improve the SOH estimation accuracy.

3.1.3. RUL prediction

RUL prediction is critical for retired LIBs evaluation, which is the estimation of the remaining cycle time or cycle number of LIBs on the premise that the batteries can still meet the requirements of the specific application scenario [52]. Battery degrades during both storage (calendar life) and cycling operation (cycling life). Calendar life is mainly affected by SOC and temperature, while cycle life is affected by factors such as temperature, number of cycles, current rate, voltage range and DOD. Multiple methods such as empirical methods, model-based methods, artificial intelligence (AI) methods, and hybrid methods, have been proposed for RUL prediction [60,61].

Lyu et al. [62] proposed a novel EM-based PF framework to predict RUL, in which state variables such as electrolyte volume fraction, specific interfacial area and exchange current density were selected from internal SOH-related features. The methodology combined the electrochemical mechanism of the battery with data-driven PF method, and achieved RUL prediction with high accuracy and high robustness for different types of batteries. When the beginning of prediction (BOP) was 50 cycles, the RUL prediction error was about 15.5%, and when the BOP was 80 cycles, the error was less than 5%. This showed that using more historical information could lead to more accurate prediction results. The main challenge of the EM-based RUL prediction approaches is the need to fully understand the electrochemical degradation process inside the LIBs. Garg et al. [63] proposed a coupled stress-AI method to analyze

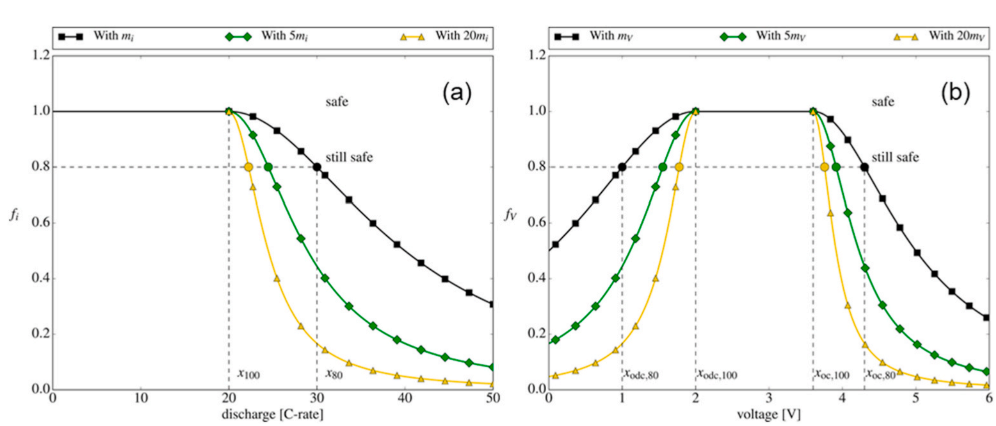

Fig. 3. SOS relationship with different abuse conditions [50]. (a) different current rate conditions. (b) voltage abuse including undervoltage and overvoltage.

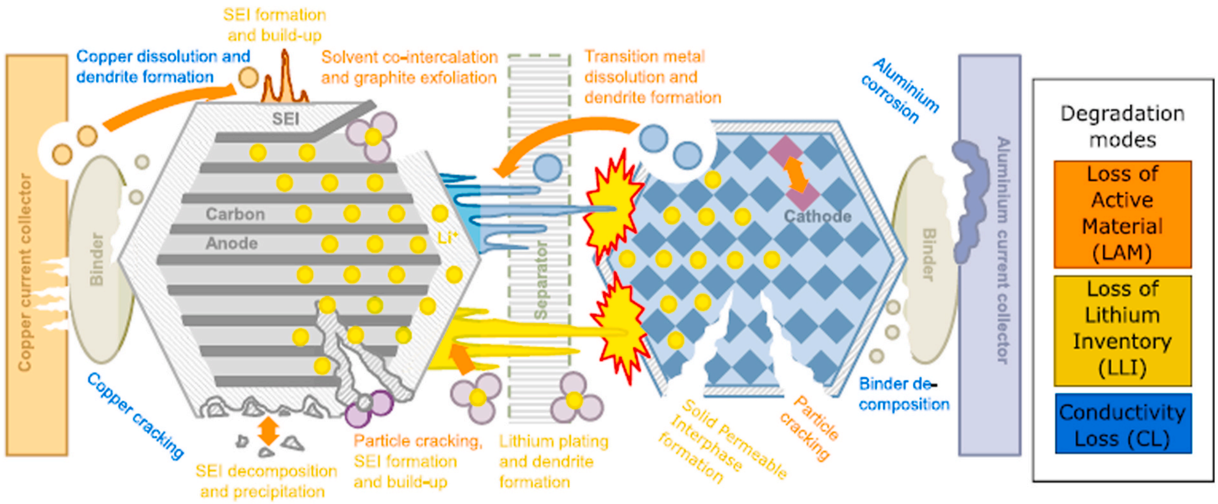

Fig. 4. Major side reactions in LIBs that cause performance degradation [51].

the residual energy (remaining life) for retired EV batteries, where genetic programming approach was utilized to predict the remaining capacity as a function of various applied stresses. 1510 groups of experimental data were obtained to verify the feasibility of the approach and 99.7% of the simulation results were within the experimental data range. Zhang et al. [64] developed a RUL prediction method using random forest (RF) and recursive least square (RLS) algorithm. The RF algorithm was used to identify the battery aging stage, and the RLS algorithm with forgetting factor was adopted to predict the EOL cycle based on aging stage. The calculation could be completed in 0.3 s. When the first 300 cycles of test data were used for model training, most of the prediction errors were less than 10 cycles. The experimental results showed that the proposed RUL prediction method can achieve high accuracy with low computation burden.

3.2. Screening

Screening approaches in the remanufacturing process can help identify the degraded batteries within the EOL pack, which will then be replaced with qualified cells. The screening process can be performed based on differences in cell parameters or properties, or based on data-driven methods.

Liao et al. [65] argued that the visual inspection and capacity measurement were not enough to achieve a satisfactory classification of retired EV LIBs, and proposed a screening method based on capacity value and electrochemical impedance spectroscopy (EIS) characteristics. Experimental data demonstrated the correlation between Warburg impedance, charge transfer resistance and the battery's capacity. Finally, 20 batteries in good conditions were selected from 60 retired batteries based on the comprehensive comparison. Jiang et al. [66] studied the consistency and aging characteristics of retired LFP batteries based on ICA method, and concluded that the loss of Li inventory was the dominant cause for capacity degradation. Quantitative analysis results showed that the changes in IC peaks and peak areas could represent the variations of anode materials, lithium inventories and reaction kinetics, therefore ICA result could be used for high-precision screening strategies.

3.3. Disassembly, reassembly and test

Disassembly steps include opening the battery framework, removing the electrical and mechanical connections between the components, as well as removing auxiliary electronic parts [7,67]. But until now there is no standardization for the configurations and structures of the battery packs and modules, and the disassembly of battery systems still requires human participation and needs to be handled manually [68]. Due to the high pack voltage, the disassembly process is usually dangerous, requiring skilled technicians and specialized equipment [8], while robotic battery disassembly could decrease the risk of human injury and reduce production cost [69]. Virtual disassembly can realize simulated disassemble based on the necessary information such as the system structure, components, connections, weights, and materials. Therefore, it is suitable for outlining different disassembly possibilities to optimize the disassembly steps [70]. Another advanced technology for battery disassembly is based on cloud platform, which can collect field information through the internet, and arrange the disassembly procedure according to the cloud computing results [71]. With the help of cloud modeling and cloud computing, the net joint movement of the robot could be reduced by more than 50%.

The reassembly process is very similar to disassembly steps. Because the remanufactured packs are used for the same purpose, the original test methods are still applicable. In addition, the original battery management system (BMS), thermal management system (TMS), and equalization management system (EMS) usually do not need additional modifications.

3.4. Applications of remanufacturing

Global Battery Solutions (formerly Sybesma's Electronics) developed a "cut-and-paste" remanufacturing method, which can repair defective packs by diagnosing, removing defective cells, and replacing the vacancy with healthy ones [72]. US-based Spiers New Technologies (SNT) can provide 4R services (repair, remanufacturing, refurbishing, and repurposing) for EV packs used by OEMs such as Nissan and General Motors. SNT also plans to provide remanufacturing services to the European and China automotive market [4,73]. In Namie, Japan, Nissan has built a facility to remanufacture EV batteries for Leaf owners with a cost of about $2850 (24 kWh) [74].

Though remanufacturing can save about 40% cost compared to new packs, currently there is no large-scale remanufacturing applications [75]. Because each OEM has its specific battery chemistry, cell type, module structure, and pack solution, it will be difficult to replace defective batteries with qualified cells from other OEMs. In addition, safety issues during the inspection, pack disassembly and reassembly, and cell replacement require professional approaches, skilled technicians, dedicated equipment and specialized workbench [15]. The responsibility and legal concern of the remanufacturing are other challenges that the large-scale industry applications have to overcome [76].

4. Repurpose

Repurpose is similar to remanufacturing, especially in terms of evaluation methods, screening methods, and disassembly processes. However, the pack structure may change, new software and/or hardware for BMS may be applied [77], and different TMS and EMS can be used because the repurposed LIBs will begin their second life in different scenarios. The pack will be disassembled into modules (cells in EV applications are usually connected by welding approach) and regrouped based on the screening results, during which necessary refurbishment may be applied [36,78].

4.1. Regroup algorithms

If retired batteries with great inconsistencies are grouped together, the risk of over-charging, over discharging, accelerated aging, and even thermal runaway will increase [6]. Therefore, it is essential to regroup the batteries with similar characteristics such as capacity, internal resistance and open circuit voltage (OCV).

Many algorithms have been proposed for LIBs regroup, including parameter-based methods and dynamic feature-based methods [79].

Huang et al. [79] proposed a battery regrouping method based on feature extraction and clustering methods. The proposed method can extract the features of the battery discharge time series, search the density peaks, and then cluster the batteries with improved k-means algorithm. The experimental results showed that compared with the traditional k-means method, the improved method could achieve similar performance and reduce the convergence time by about 79%. Xu et al. [6] proposed a clustering method for retired LIBs regrouping based on traversal optimization, which could form clusters with high differences between clusters and low variations within clusters. Experimental results showed that the proposed regrouping method were more effective than k-means algorithm and affinity propagation method, and were applicable for large samples applications. Zhou et al. [80] proposed a rapid classification method for retired EV LIBs, which could regroup the batteries depending on capacity and internal resistance with the help of piecewise linear fitting and radial basis function neural network (RBFNN) methods. The data set was collected by piecewise linear fitting method, and then trained by RBFNN for large-scale LIBs classification. 4 Cells with similar characteristics were selected from 12 retired batteries and connected in series to form a pack. At the end of discharge, the voltage difference of the pack was about 34 mV, and the root mean square error (RMSE) was about 15.5 mV. Experimental results proved that the proposed regroup method could obtain satisfactory consistencies.

4.2. Battery management functions

Battery states estimation, such as SOC, state of power (SOP) and SOH are key management issues for the safety, reliability, and efficiency of EV packs [81]. So far, most studies on battery states estimation have focused on EVs, while relatively few on echelon use scenarios.

4.2.1. SOC estimation methods

Various methods for implementing SOC estimation have been proposed, which can be roughly divided into three main categories, including direct calculation, model-based [82] and data-driven methods [83]. The direct calculation method can obtain the initial SOC based on open circuit voltage (OCV), and calculate the SOC change by coulomb counting method. However, it has the disadvantages of cumulative error and lack of adaptive capabilities. ECM-based methods are widely adopted due to the merits of simple structure and good scalability, but the model parameters need to be adjusted according to operating conditions. Data-driven methods have the potential to be widely adopted due to the flexibility and adaptability, while they require high-quality training dataset and may become over-fitting in practical applications.

Considering the different charging and discharging efficiencies caused by degradation, Cusenza et al. [84] proposed a simplified SOC estimation method for ESS made from retired EV LIBs. Similar to coulomb counting, the simplified method can estimate SOC based on energy flows integration. Tong et al. [85] repurposed the retired LFP cells as a household energy source, and proposed a worst-difference extended Kalman filter (EKF) method based on ECM for SOC estimation. The experimental results show that when used in cooperation with a PV array, the system can reduce grid energy consumption by 81%. Yang et al. [86] developed a "special and difference" model to describe the imbalance of the aged battery pack. With the help of multi-time scale EKF method based on second order ECM, the SOC of the special battery, remaining cells and the pack could be estimated on different time scales. Under New European Driving Cycle (NEDC) and Urban Dynamometer Driving Schedule (UDDS) test conditions, the maximum error of SOC estimation was about 4.6% and 3.9% respectively, which proved the feasibility of the proposed method.

4.2.2. SOP estimation methods

Accurate SOP estimation can avoid overcharging or over discharging abuse of the pack, thereby ensuring the best performance of the battery system. SOP estimation methods can be divided into characteristic map (CM)-based methods and model-based methods [81]. CM-based methods rely on the relationship between SOP and battery characteristics such as SOC, temperature, SOH, and time duration, which can be obtained through offline testing. With the help of advanced control algorithms, ECMs are popular for the online SOP estimation under dynamic conditions. However, the generalization ability of model-based methods is usually not satisfactory.

Shen et al. [87] compared the accuracy and efficiency of various peak power test methods including hybrid pulse power characterization (HPPC), Japan Electric Vehicle Standard (JEVS), and constant current, and proposed an optimized JEVS method by alternative charging and discharging process. The experimental results showed that within the range of 10%–95% SOC, the peak discharge current error of the JEVS method was less than 10%, which validated the accuracy of the improved method. Wei et al. [88] developed a novel parameter identification method based on first order ECM with hysteresis. The parameters can be obtained online with the help of extreme seeking algorithm and the SOP can be then estimated using the voltage and current limitations. The simulation results on experimental data prove the feasibility and stability of the proposed parameter identification and SOP estimation methods. Zhou et al. [89] proposed an EKF-based SOP estimation method for series-connected batteries with low computational cost, which could select the representative batteries depending on the variation of characteristic voltages and ohmic resistances. The experimental results under dynamic condition at different temperatures proved the accuracy, feasibility and robustness of the proposed estimation method.

4.2.3. SOH estimation methods

Accurate SOH estimation, which can support fault diagnosis, SOC and SOP estimations, is a key issue to guarantee the safety, reliability and efficiency of the battery system [45]. When integrated into the BMS, SOH estimation methods must overcome the challenges of computational cost and parameter identification.

Li et al. [90] proposed a SOH estimation method for NMC LIBs based on the ICA approach. Gaussian filtering method was used to smooth the IC curves without deforming the feature range, and the relationship between the variation in features range and battery capacity fading was used to identify the battery SOH. The experimental results showed that the maximum absolute error of SOH estimation was less than 2.5%, which validated the feasibility and generalization of the SOH estimation method. Zhang et al. [91] discussed the capacity performance of 24 retired LFP modules using 6 different capacity test protocols, and concluded that the constant current charge of 1/3C was the best trade-off between capacity accuracy and test duration. The experimental results show that module-level reuse may have greater value than pack-level reuse due to the inconsistencies between modules. Li et al. [92] designed a variant long-short-term memory (LSTM) NN based SOH estimation method, which can obtain the old data and new information at the same time and extract more useful features. The experimental results on NASA dataset verify the accuracy of the method with 2.16% average root mean square error.

4.2.4. Joint state estimation

Battery states such as SOC, SOP, and SOH are strongly coupled and interact with each other in EV and reuse applications. Therefore, multi-state joint estimation methods integrated in BMS have the potential to obtain high accuracy and high robustness [45].

Shen et al. [93] proposed a SOC, SOH and SOP co-estimation method, which can achieve real-time estimation at the same time. SOC can be estimated by EKF algorithm based on a second-order ECM, and SOP and SOH related parameters can be obtained using the improved RLS algorithm. With the updated capacity from SOH estimation, the accuracy of SOC estimation could be significantly improved. Under dynamic stress test (DST) and Federal Urban Driving Schedule (FUDS) test conditions, the estimation errors of SOH and SOC were 0.5% and 1.1% respectively, which proved the effectiveness and accuracy of the proposed method. Hu et al. [94] developed a hierarchical framework for SOC, SOH and SOP co-estimation at multiple time scales. The framework included four parts: real-time SOC estimation based on a modified moving horizon method, periodic model parameters updates, offline SOH estimation based on coulomb counting, and periodic calculation of SOP. Experimental results verified the feasibility of the proposed method with an SOC estimation error less than 3%, a voltage error within 25 mv, and a capacity error less than 3% respectively. Our group [95] proposed an interactional hierarchical management strategy, namely cyber hierarchy and interactional network (CHAIN). With the help of CHAIN, BMS can realize real-time monitoring and upload critical data to the cloud server simultaneously, then the virtual battery model with high fidelity to the real world can be constructed on the cloud, thereby achieving a balance between estimation accuracy and model complexity.

4.3. Thermal management functions

LIBs are sensitive to temperature and usually achieve the best performance near room temperature. TMS can maintain the maximum temperature and temperature variance within an appropriate range, thereby ensuring the health and safety of the battery system. Mediums used for battery cooling include air, liquid, phase change material (PCM), and heat pipes, etc. According to whether the medium is in contact with the battery, TMS can be divided into direct cooling and indirect cooling systems. Depending on whether power consumption is needed, TMS can be divided into active and passive management systems [96].

Compared with new batteries, repurposed LIBs will suffer from degradation of safety and reliability performance, which requires more attention on the design and implementation of TMS. TMS should be selected based on a comprehensive consideration of factors such as temperature performance, cost, space, efficiency, and reliability [97].

Bai et al. [98] presented a TMS solution based on heating plate and PCM for outdoor 48V battery pack, and the combination of heating and preservation process could help maintain the pack temperature in optimum range for several days. The simulation result showed that when the heating plate was 0.16 m^2 and placed horizontally under the pack, the maximum temperature difference between the batteries was less than 5 °C. Compared to other TMS with different size and arrangement, the temperature homogeneity was improved effectively. White et al. [99] compared the performance of various retired EV LIBs in grid frequency regulation (FR) applications. Experimental results showed that the thermal characteristics of second life batteries were greatly affected by TMS and working conditions. Under aggressive FR conditions, the peak temperature of Lishen and Leaf batteries (with passive TMS) reached 48.7 °C and 49.6 °C respectively, while the peak temperature of batteries with active TMS (such as Tesla and Volt) was below 40 °C. In addition, active TMS could achieve better heat accumulation and temperature fluctuation performance, thereby were more suitable for FR applications. When the requirements such as temperature range, thermal response, and space constraints can be met, the original TMS of retired LIBs can be retained, otherwise, a specific TMS should be designed and implemented.

4.4. Equalization management functions

After hundreds of charge-discharge cycles in EV applications, the battery cells in the same pack will develop different capacity and internal resistance. Such imbalance may exacerbate and lead to high risk of system failure and safety issues during reuse period. EMS can improve cell balances, maintain consistent operating conditions and reduce maintenance costs [44]. According to whether excess energy is dissipated, EMS can be divided into active mode and passive mode systems.

Active equalization is widely studied due to the high efficiency and high balancing capacity. Ma et al. [100] proposed an active balancing circuit and corresponding online equalization strategy for the repurpose of LMO batteries from pure electric buses in load shift ESS applications. According to the inconsistency evaluation of capacity and internal resistance among the retired batteries, equalization approach was adopted based on the capacity utilization with a maximum balancing current of 2A. The feasibility and reliability of the online balancing system were verified on the test bench, and the capacity utilization was increased by 9%. Bat-Orgil et al. [101] developed an active equalizer for retired nickel-metal batteries from hybrid electric vehicles, which can transfer excessive energy from high-SOC cell to low-SOC cell with the help of generation control circuits. Simulation and experimental results showed that the equalization circuit could effectively balance the series-connected batteries during the discharging process. Ma et al. [102] proposed an active SOH equalization method for ESS using retired batteries based on bi-directional flyback circuits. Batteries with higher SOH will have deeper DOD than the batteries with lower SOH, which can improve the SOH inconsistency of batteries within a string. Simulation models and experimental prototype bench were developed to verify the effectiveness of the active SOH balancing method. To improve the balancing speed of passive equalization, ceramic load resistors were adopted as balancing components in a maintenance-free ESS [103], which had a balancing ability up to 1.08 A and could fulfil the equalization requirement of 320 Ah batteries with significant imbalance. The off-grid ESS with high reliability had been applied on an island in Tanzania.

4.5. Applications of repurpose

Repurposed batteries can be utilized in less demanding scenarios including ESS, UPS, electric scooters, and electric forklifts. According to the difference in energy capacity, the repurposed applications can be classified as industrial, commercial and household grade applications [104]. Based on mobility, repurposed solutions can be divided into stationary level (grid-connected ESS, green buildings, etc.), quasi-stationary level (such as ESS for major events), and mobile level (power supply in e-scooter or forklift) [105]. ESS scenarios such as smart grid, telecom backup towers and buildings, are probably the most popular application scenarios for reused EV LIBs, where similar modules are screened and repurposed to satisfy the requirement of different scenarios [38]. Some typical applications are shown in Table 2.

4.5.1. Grid related applications

When used in on-grid applications, the second-life batteries have the potential to achieve peak shaving and load shifting by charging during

Table 2
Typical application cases for purposed EOL LIBs.

Applications		Mobility	Related Company	Capacity
	Energy storage system in John Cruyff Arena [4]	Stationary	Nissan, Eaton, etc	4 MWh/ 4 MW
	Backup supply for telecom tower in China [78]	Stationary	China Tower	30 kWh
	EV Charger in California [106]	Quasi-stationary	Siemens, Freewire	80 kWh/ 11 kW
	Delivery vehicles in China [78]	mobile	SF Express	2-3 kWh

off-peak period and output buffer energy during periods of high energy demand [107,108]. ESS can also be used to maintain the grid stability by voltage or frequency regulations [109]. Matsuda et al. [110] studied the possibility of reusing EV battery for renewable energy power system in remote island to solve the problems of frequency fluctuation and power surplus, and evaluated the necessary battery requirement such as power and capacity based on real data measured from Koshiki Island for about 7 months. Tong et al. [111] proposed a residential energy solution based on repurposed batteries and PV arrays to reduce daily grid energy consumption. The experimental data showed that the solution could effectively alleviate the contradiction between solar intermittency and fluctuated energy demand, and reduce the grid consumption by 64%–100%. Debnath et al. [112] proposed a smart grid system model that could congregate grid-able EVs and repurposed batteries together to serve as backup systems for the grid. Simulation results showed that such model could save 70% of the cost compared with conventional solutions, and could recover the investment for the second-life batteries in about 1.5 years.

General Motors has partnered with ABB to repurpose batteries from Chevrolet Volt as grid-based energy storage devices that can store off-peak energy in the night and transfer it back at appropriate time [76]. In the John Cruyff Arena in Amsterdam, 280 Nissan Leaf battery modules are used to form a backup power system, which is the largest ESS assembled by the repurposed LIBs in Europe with 4 MW nominal power and 4 MWh nominal capacity [4,106]. The power system can obtain energy from PV system during the day or from grid energy at night with a low cost.

4.5.2. Commercial grade applications

Repurposed batteries can serve as backup energy for EV charging, data centers, hospitals, and telecom towers [104]. China Tower, the world's largest telecom infrastructure service provider with 2 million telecom tower base stations, has signed partnership agreement with 16 automotive OEMs and battery manufactures, and has stopped purchasing lead-acid batteries since 2018. The company plans to replace the lead-acid cells with repurposed LIBs gradually. Considering that the energy capacity of a single tower backup system is about 30 kWh, a total of 2 million spent EV LIB modules can be repurposed in the next few years [78]. California-based FreeWire Technologies has partnered with Siemens to commercialize a cloud-based mobile EV charging station called Mobi Charger, which is powered by repurposed Leaf batteries. The Mobi Charger can be easily moved to provide EV charging and can charge five cars per day [106].

4.5.3. Other scenarios

Retired LIBs can also be reused in homes and buildings, industrial vehicles (such as forklifts, cranes, transport trolleys and sweepers) and minor motorized EVs (such as electric bicycles, electric scooters and golf carts) with lower performance requirements [105,106].

For instance, in China, due to the advantages of low cost, small size and long service life, thousands of express delivery vehicles powered by repurposed EOL LIBs have been used by SF Express and some other express delivery companies [78]. Nissan and Eaton have jointly launched a residential energy storage system called xStorage, which combines second-life batteries from Nissan Leaf and converters from Eaton. The xStorages system can effectively reduce energy cost based on time-sharing pricing [106].

5. Recycling

Recycling is beneficial because valuable materials in LIBs become part of the circular value chain, thereby partially alleviating the requirement for raw resources [12]. Due to the high scalability and ease of handling, recycling is considered as the most widely applicable solution for EOL LIBs [4].

The recycling of LIBs is still quite complicated due to the complex pack configuration, diverse battery shapes and various battery chemistry selections [113]. Firstly, in addition to multiple modules, LIB packs also composed of BMS, TMS, flame retardant, insulation, support, connection, fastening, sealing and other functional components, and the architecture of packs varies greatly between different OEMs [114], which increases the difficulty of recycling. The differences in battery shape also affect the difficulty of recycling; compared to cylindrical batteries, prismatic batteries and pouch cells are relatively easy to be separated and recycled due to their planar electrode structure [7]. In addition, the active materials in cells vary with manufacturers. Most LIBs use graphite as anode, and some use lithium titanium oxide (LTO). Cathode materials mainly include olivine-type (LFP), layer-type (LCO, NMC, NCA) and spinel-type (LMO) structures, and currently, LFP and NMC family are the most widely used cathode materials [115]. LFP has the advantages of high thermal stability and low production cost, but the low energy density limits its application [116]. NMC battery is widely adopted in EV market due to its superior electrochemical performance, but the energy density still cannot meet the mileage requirements of the customers [117]. Nickel-rich cathode materials such as NMC811 are researched to further increase the energy density of LIBs [118]. Recycling more materials (such as foils and electrolytes) and pack components (such as BMS and TMS) can improve the recovery rate and profit [119]. However, the diversity of materials will further increase the technical difficulty and cost of battery recycling.

Current recycle interests mainly focused on the recycling of valuable metals, such as Co, from cathode materials of spent LIBs [120]. Although the recycling process is very complicated, it generally consists of two stages. The first stage is pre-treatment, includes discharging, disassembly, crushing, screening and separation, which removes cells from EV packs and then break down cells into valuable particles that can be used in the next process [7,28,121,122]. Materials extraction is the second stage, which mainly includes pyrometallurgical, hydrometallurgical, bio-hydrometallurgical and direct recycling [123]. In addition, valuable resources in electrolyte and anode materials can also be recycled [124,125].

5.1. Pre-treatment

The pre-treatment stage is to separate and enrich valuable components and materials from EOL LIBs based on the differences in physical properties (such as shape, density and magnetic properties). Pre-treatment can help increase recovery rate, reduce energy consumption, avoid safety risks and reduce environmental hazards [121,122]. Pre-treatment usually includes various chemical and physical operations, such as stabilization, heat treatments, vacuum pyrolysis, milling, and sieving [126,127]. Generally, the main procedure of pre-treatment can be divided into discharging, disassembly, crushing, and separation processes, which is shown in Fig. 5 [122,128].

5.1.1. Discharging

Spent LIBs still have residual energy which may lead to short circuit and even explosion during pre-treatment process, therefore, the batteries usually need to be stabilized first [129]. The most popular stabilization methods are brine method and Ohmic discharge [130].

The brine method soaks the used LIBs in solution such as NaCl and Na_2SO_4 for a sufficient time duration to completely discharge. Nie et al. [131] immersed the LCO batteries in Na_2SO_4 saturated solution for 24 h to achieve deep discharge. The advantage of the brine method is that the remaining energy can be completely discharged by short circuit without overheating during the process. The immersion process usually takes a long time, and the brine method is not suitable for high voltage packs, which may cause high rate of electrolysis and release vigorous gases [7].

The Ohmic discharge method refers to discharging the battery with an external load circuit. Kruger et al. [132] discharged the NMC batteries with resistances and then recycle the active materials. He et al. [125] discharged 24 series-connected cells by connecting to the load, and the batteries could be discharged to 2 V synchronously within 2 h. During the process, a BMS with temperature sensors was used to monitor the pack status. Compared with the brine method, ohmic method can reduce the time to fully discharge the spent LIBs. But LIBs will suffer from a large amount of heat and high temperature in a short time, which may further lead to the risk of thermal runaway.

5.1.2. Disassembly

Disassembly is a necessary procedure in the pre-treatment process of EOL LIBs. As shown in Fig. 6 [27], the structure of the battery systems can be very complicated. In the disassembly process, the battery system will be dismantled into module level or cell level, and the steps include opening the battery framework, cutting the electrical connections between the components, removing the mechanical connections between the components and the base, as well as removing auxiliary electronic parts [7,67]. Until now there is no widely accepted standard for the configurations and structures of battery packs and modules, therefore, there could be huge differences between various EV packs. At present, the disassembly of battery packs still requires human participation and needs to be handled manually [68].

The objective of disassembly process is to minimize environmental costs and maximize economic benefits [133], and many advanced technologies have been used for LIB disassembly. As described above, robotic assistance, virtual disassembly, and cloud-based technique may be the tendency of disassembly technologies.

5.1.3. Crushing

After disassembly process, the LIBs need coarse shredding or fine crushing to reduce the granularity of the materials for further recycling [134]. The battery shredding or crushing process can be executed under an inert gas environment such as carbon dioxide to prevent the risk of thermal runaway and reduce pollution [135]. The LIBs can also be crushed in lithium brine to neutralize the electrolyte and prevent gas emissions [4].

Wang et al. [136] performed the mechanical shredding process by a granulator under a fume hood. Experimental results showed that the pieces with size greater than 6 mm were mostly casings and separators, and the graphite anode and the cathode active materials formed the majority of the pieces with size less than 0.5 mm. The active materials can be segregated into different size fractions with the help of shredding and crushing. Zhang et al. [137] crushed the discharged spent LIBs on an impact crusher at a speed of 3000 rad/min for 20 s, and then sieved the crushed material to separate the coarse product with a size of +0.2 mm and the fine with a size of −0.2 mm. Studies demonstrated that the coarse products were mainly formed by metallic shell, collector foils and organic materials, and the fine products mainly consisted of graphite and cathode particles, which could be recycled in subsequent metallurgical process.

5.1.4. Separation

After shredding and crushing process, the various materials are procedurally separated and processed for further treatment steps [134]. Physical processes can be used to separate various materials based on the differences in physical properties such as size, density, ferromagnetism and hydrophobicity [135].

Wang et al. [136] performed the size-based separation process with a vibration machine and sorting sieves. The shredded LIB material was separated into five size fractions, and the ultrafine part (<0.5 mm) was mainly composed of active materials from the electrode, accounting for about 19.5% of the total weight. The proposed separation process could effectively separate materials, thereby improving the purity of the products, and had the potential for grid-scale application. Yu et al. [138] proposed a flotation-based separation process to separate LCO and graphite materials from crushed LIBs. The obvious difference in wettability among the electrode materials contributes to a concentrate grade of 97.13% and 73.56% respectively. Grinding flotation has the advantages of high efficiency and pollution free, which can facilitate industrial application. Shin et al. [139] utilized a series of separation processes to produce concentrated LCO particles. Firstly, the vibration sieving was adopted to separate large pieces of materials such as plastic and copper, then a magnetic separator was used to remove steel casing, finally the second vibration sieving was applied to eliminate small pieces of aluminum completely.

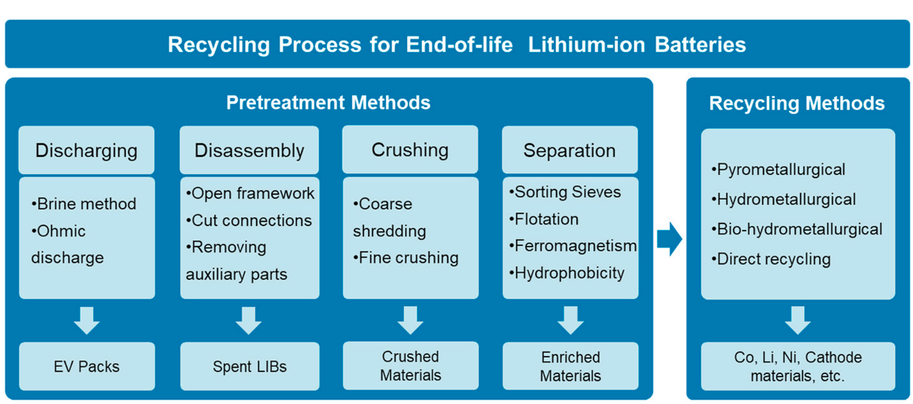

Fig. 5. Main procedure of EV LIBs recycling process [122,128].

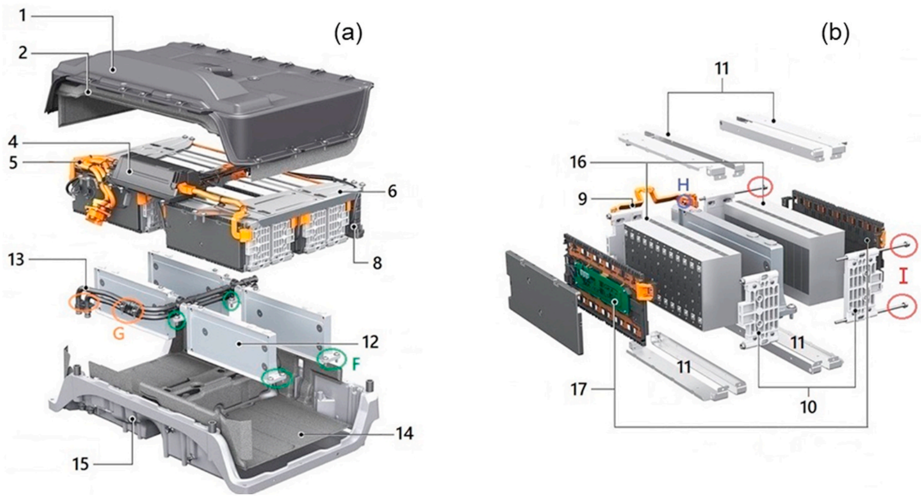

Fig. 6. Exploded view of the battery system and the module [27]. (a) Exploded view of the battery system. (b) Exploded view of the module.
1-Upper housing shell, 2-Upper insulator, 3-Plug-in cable, 4-Battery Junction Box, 5-High Voltage cables and connectors, 6-Top transverse cover, 7-Plug-in cable, 8-Battery Management Controller, 9-Module connector, 10-Side module junction, 11-Top and bottom fastener of module,12-Cooling plate, 13-Cooling pipe, 14-Lower insulation, 15-Lower housing shell, 16-Module, 17-Cell Management Controller.

5.1.5. Summary of pre-treatment

The pre-treatment process can enrich valuable components and materials such as Li, Co, and Mn, and is a necessary step for EOL LIBs recycling. The development trend of pre-treatment technologies is to improve efficiency, reduce cost and increase the purity of materials.

During the pre-treatment process, stabilization method such as discharging should be performed to avoid thermal runaway or even explosion. The disassembly step still needs human participation due to the diverse battery structures from different OEMs, and the technologies including robotic and virtual disassembly can help improve efficiency and avoid human injuries. Crushing and separation steps can contribute to enriching the valuable materials, and the multistage processes have the potential for large-scale applications.

5.2. Metallurgical technologies

Various materials such as Co, Li, Ni, Cu, Al, and phosphorus can be obtained from spent LIBs. Since the value of cathode materials account for about 40% of the total value of the LIBs, the current recycling process is mainly focused on high value metals in cathode materials, such as Co, Li and Ni [4,140]. The valuable part of materials after pre-treatment process will be sent to refining process, which includes pyrometallurgical, hydrometallurgical, bio-hydrometallurgical, and direct recycling [141,142]. The main products of the recycling process can be pure metals, alloys, compounds, solutions containing metal ions and slag [121,143].

Pyrometallurgical technology uses thermal treatment to reduce the component metal oxides to an alloy and is broadly used in the industrial recycling of EOL LIBs due to the relatively simple operation and high processing capacity [140,144]. However, the high energy consumption and low recovery rate greatly limit their application. Hydrometallurgical technology is an efficient method to dissolve and extract valuable metals from aqueous mediums [145]. Hydrometallurgical method has the advantages of low energy consumption and high recovery purity, however, the process is rather complicated [121]. Bio-hydrometallurgical technology usually utilizes the acids produced from microbial metabolization to leach valuable materials from spent LIBs [123], and may be complementary to conventional pyrometallurgical or hydrometallurgical methods due to the environmental friendliness [4,28]. Direct recycling refers to recovering battery materials without decomposing the original compound structure [146]. The direct recycling method has the advantages of low cost and low energy consumption, but may need to tailor the production process for specific material formulations [7].

5.2.1. Pyrometallurgical

Pyrometallurgical process can treat the spent LIBs just like ore. In the process, the EOL LIBs are smelted to reduce the valuable metals to a product of mixed alloy [147]. Generally speaking, pyrometallurgical process can be divided into three stages: pre-heating, plastic burning, and valuable metals reducing [144]. In the pre-heating stage, the electrolyte evaporates slowly and can reduce the explosion risk. Next, organic materials such as plastics are burned away which can help maintain the high temperature. The above two steps can be collectively referred to as thermal treatment [122]. Then the materials are smelted and reduced with the production of alloys with valuable metals such as Cu, Fe, Co and Ni, as well as slags containing Li, Al and Ca [113,148].

Pyrometallurgy method usually recovers valuable materials by reduction reactions under high temperature [122]. Due to the consideration of energy consumption, now most recycle methods aim to reduce the temperature of traditional pyrometallurgical processes to 500–1000 °C [130].

Nie et al. [131] proposed a pyrometallurgy method for LTO batteries recycling. In the thermal pre-treatment stage, the smashed materials were roasted at 400 °C for 1 h and at 800 °C for 2 h separately. Then the powder was mixed with supplementary Li_2CO_3 and calcined at 850–950 °C for 12 h to obtain $LiCoO_2$. The proposed method had the advantages of simple process and environmental friendliness, and the physical and electrochemical properties of the $LiCoO_2$ products could meet the requirements for commercial application. Li et al. [149] proposed a pyrometallurgy method to recycle valuable materials in situ from electrode materials including $LiCoO_2$ and graphite. The mixture of Co, Li_2CO_3 and graphite could be obtained by calcining at 1000 °C for 30 min, and the recovery rate of Co, Li and graphite could be 95.72%, 98.93%, and 91.05% respectively. Pyrometallurgy method with vacuum carbon reduction is a clean and energy saving technology [150]. Xiao et al. [151,152] proposed a vacuum metallurgy approach, which could recycle spent LIBs with different cathode materials. In a vacuum degree of below 1000 Pa, the binder was decomposed into gas at 300 °C for 45 min. The mixed electrode materials including graphite and LMO were heated at 800 °C for 45 min, and the optimal recovered rate of Li is 91.3%, the purity rate of Mn_3O_4 was 95.11%, and the purity rate of Li_2CO_3 was 99.7%.

Pyrometallurgical approach is the most popular method used in the industry for the recycling of Ni–Cd, Ni-MH and LIBs, which can smelt and reduce valuable materials at high temperature to produce alloy and metal oxides [153]. Pyrometallurgical method has the advantages of mature process and the sorting pre-treatment are not necessary. However, the disadvantages of pyrometallurgical method are the high energy consumption, hazardous gas emission, and that the output alloy usually needs further processing [130,140]. Furthermore, for batteries that do not contain Ni or Co (such as LFP), the economics of such method may not be satisfactory [154].

5.2.2. Hydrometallurgical

Hydrometallurgy takes advantage of aqueous solutions to leach the valuable metals from EOL LIBs. Due to the merits of low energy consumption and high extraction rate, the hydrometallurgical process has become the research focus for the EOL LIBs recycling [120,155]. The keys procedures of the hydrometallurgy process are leaching, precipitation and solvent extraction. Leaching means the dissolution of target active materials by leaching agents. Typical leaching agents include inorganic acids, organic acids, and alkaline solutions [130,140], while chelating agents can also be adopted as the leaching agent [156]. Inorganic acids such as HCl, HNO_3 and H_2SO_4, and strong organic acids such as citric acid, ascorbic acid and oxalic acid can be used as the leaching agents for hydrometallurgical process [157,158], while reducing agents such as H_2O_2 are often used to help dissolve of active materials [130,141,154]. Then treatment such as precipitation, ion exchange, solvent extraction, and electrolysis can be used to remove impurities or separate metals [4,159].

Li et al. [160] proposed a hydrometallurgy process to leach Co and Li from spent LCO batteries, and compared the leaching efficiency between different acids. The experimental results show that citric acid with H_2O_2 as additional reductant can leach more than 96% Co and almost 100% Li, which is superior to HCl and H_2SO_4 in terms of efficiency and environmental friendliness. Chen et al. [120] proposed a gradient process to regenerate cathode materials from spent NMC. Firstly 99.7% Li was selectively extracted under optimized tartaric acidic conditions at 80 °C for 30 min, and then transition metals (Ni, Co and Mn) enriched residues were completely leached in sulfuric acidic medium at 70 °C for 30 min. Next, the transition metals could be sufficiently precipitated by Na_2CO_3 and $NH_3{\cdot}H_2O$ solution. The gradient process can minimize the consumption of acids and achieve a balance between recycling efficiency and environmental impact. Li et al. [161] proposed an environmentally friendly leaching process to recover Li from LFP batteries with low cost. Under the specified condition (oxalic acid concentration of 0.3 mol/L, solid/liquid ratio of 60 g/L, reaction time of 60 min, and reaction temperate of 80 °C), the proposed method could extract about 98% of Li and 92% of Fe (in terms of $FeC_2O_4{\cdot}2H_2O$).

Although the acid leaching methods have the advantages of strong leaching ability and high efficiency, they have the disadvantages of toxic gas emissions and weak leaching selectivity. To address this problem, the ammoniacal alkaline solutions have been used on the leaching of valuable metals such as Li, Co, and Ni because of the low toxicity, high selectivity, and evaporative recyclability [162–164].

Zheng et al. proposed [162] a two-step ammonia leaching process to recycle Ni, Co and Li from cathode scrap provided by a recycling company. After pre-treatment, the cathode powder was immersed into the ammonia and ammonium sulfate leaching reagents. Under the optimum conditions at 80 °C for 5 h, the total leaching rate of Ni, Co and Li was more than 98% separately, however, the selectivity rate of Mn was only 6.34%. Qi et al. [164] proposed an effective ammonia leaching process to recover valuable metals from waste LIBs. Under the optimal conditions (the concentration of $NH_3{\cdot}H_2O$ and NH_4HCO_3 was 120 g/L and 75 g/L respectively at 80 °C for 4 h), the process yielded a leaching rate of 91.16% and 97.57% for Co and Li respectively. Wang et al. [163] compared the effects of various species of ammonia salts, ammonium salts and reductants on the leaching of Li, Co, Ni from spent LIBs. The increase of the electrode potential of the reductant could effectively accelerate the selective leaching reaction. When using NH_3–NH_4Cl solutions mixed with $(NH_4)_2SO_3$ as a reductant, leaching process could extract 100% Co, 98.3% Ni and 90.3% Li. And the proposed method could significantly reduce the reaction time.

5.2.3. Bio-hydrometallurgical

Bio-hydrometallurgical process (bioleaching), namely the extraction of valuable metals from waste materials by means of microbial metabolism or microbial acid production, has been adopted as an alternative method for recovering valuable materials from spent LIBs owing to the merits of high efficiency and low cost [28,165].

Biswal et al. [166] applied *Aspergillus niger* (*A.n*) strains and *Acidithiobacillus thiooxidans* (*A.t*) for the leaching of Co and Li from spent LIBs obtained from a recycling company in Singapore. After bio-hydrometallurgical process, the fungal leaching was first filtered, and precipitating agents such as sodium sulfide, sodium hydroxide, sodium oxalate, and sodium carbonate can be used to recover Co and Li. The experimental results showed that the *A.n* bioleaching method could dissolve 82% Co and 100% Li under the optimal conditions, which was more effective than the *A.t* bioleaching or acid leaching. The *A.n* bioleaching method also had the potential to recover large quantity of LIBs in an environmentally friendly way. Heydarian et al. [167] proposed a two-step bio-leaching process based on a mixture of *Acidithiobacillus ferrooxidans (A.f)* and *A.t* to recycle valuable metal from spent LIBs. Under the optimum initial conditions of 36.7 g/L concentration of iron sulfate, 5.0 g/L sulfur, pH of 1.5 and inoculum ratio 3/2 of *A.f/A.t*, the maximum recovery rate was about 99.2% Li, 50.4% Co and 89.4% Ni. Experimental results proved the effectiveness and environmental friendliness of the proposed method.

Though bioleaching methods of LIBs have the advantages of low cost and environmental friendliness, they still remain in the laboratory stage due to the drawbacks of long incubation time and easy contamination [164].

5.2.4. Direct recycling

Direct recycling is a recovery method which can recover battery materials and retain their original compound structure without decomposition [4,146,168]. Direct recycling method has the advantages of low cost and low energy consumption due to the avoidance of expensive purification steps, which can help the economical recycle of lower-value cathodes namely LMO and LFP. A majority of spent LIBs components can also be recovered by direct recycling process. However, direct recycling process may require processing of a variety of unstable feedstocks and must be tailored to specific cathode requirements [7].

Li et al. [142] proposed a direct green regeneration process to recover high purity LFP cathode materials from scrapped LFP batteries. After pre-treatment process, the recovered LFP materials were further directly regenerated with Li_2CO_3 at 600–650 °C for 1 h under Ar/H_2 gas flow. The experimental results demonstrated the low cost and feasibility of the direct regeneration process. The products performed excellent physical, chemical and electrochemical properties to meet the requirement of automotive applications. In addition, graphite anode material, Al, Cu and electrolyte solvent can also be recycled. Shi et al. [169] proposed a green energy-saving method for the regeneration of LCO cathode materials, which could retain the initial morphology and structure with outstanding electrochemical performance. The method combined hydrothermal treatment with short-time annealing on the recycled particles without complicated leaching, precipitation and waste treatment steps. It could also handle batteries with different degradation status by eliminate the compositional and structural defects [170]. Experimental results showed high potential for large-scale recovery of EOL LIBs cathodes.

5.2.5. Comparison of metallurgical technologies

Table 3 summarizes the comparison of the main recycling methods [7,8,142,166]. Pyrometallurgical methods have the advantages of simple processes and large-scale production capacity, and have been applied in industry for spent LIBs recycling. However, the high energy consumption and harmful gases emission limit the further applications. Due to the advantages of low energy consumption, high recovery purity and high extraction rate, the hydrometallurgical process has become the

Table 3
Comparison of the main recycling methods [7,8,142,166].

Recycle method	Advantages	Disadvantages
Pyrometallurgical	simple processes, high productivity, industrial-scale capacity	high-temperature, high energy consumption, hazardous gases emission
Hydrometallurgical	low energy consumption, less waste, high purity, high extraction rate,	complex steps, huge reagents consumption, waste water emission
Bioleaching	environmentally friendly, high efficiency, low cost	long time for cultivation, susceptible to contamination
Direct recycling	low pollution, low greenhouse gases emission, low energy consumption,	takes time to mature and commercialize

research focus for EOL LIBs recycling. However, this recovery methods have to overcome the problems of complicated processes, huge reagents consumption, and long processing time. Although Bioleaching methods have the merits of high efficiency, low cost and environmental friendliness, the microbial methods require long time for cultivation and can be easily contaminated. The direct recycling methods have the advantages of low cost, low energy consumption and low pollution. However, they are still in the laboratory stage and need some time to be commercialized.

Although different technologies have been researched to recover valuable metals from used LIBs, the overall recycling performance such as economy, environment, and timeliness is still unsatisfactory due to the high uncertainty of the composition of spent LIBs. Therefore, combined recycling processes can be performed [4,171]. For instance, Sun et al. [172] proposed a novel process combining vacuum pyrosis and hydrometallurgical technology, which can obtain 99% recovery efficiency of Co and Li under the optimum conditions. Umicore [173] performed a combined pyrometallurgical and hydrometallurgical recycling process to recover Co and Ni from spent LIBs in form of $LiCoO_2$ and $Ni(OH)_2$.

5.3. Recycling of anode materials and electrolyte

5.3.1. Recycling of electrolyte

Hazardous electrolyte in spent LIBs could be another valuable resource after safe and proper treatment. Several organic solvents can be used as extraction agents, while the extraction with supercritical carbon dioxide has the advantages of less solvent impurities and simple purification process [174].

Grützke et al. [175] compared the extraction behavior between supercritical CO_2 and liquid CO_2 methods. Supercritical CO_2 can achieve higher recovery rate of cyclic carbonate such as EC, while Liquid CO_2 could extract higher rate of linear carbonates and achieve higher overall recovery rate of carbonates. Additional solvents could further improve the extraction of electrolytes. The best overall recovery rate of electrolyte for a Panasonic 18,650 cell (NMC/graphite) could reach (89.1 ± 3.4) wt%. Mönnighoff et al. [176] performed the supercritical CO_2 extraction with acetonitrile as the co-solvent under the condition of 40 °C and 20 MPa. The extraction started with a static step for 30 min, and then a CO_2 flow-through step with addition of acetonitrile for 5 min. After repeating the procedure for several times, the extraction of electrolyte was completed. Liu et al. [174] developed a supercritical CO_2 extraction method to extract electrolyte from spent LIBs. The batteries were dismantled in a dry glove box filled with argon, and then transferred to a supercritical CO_2 extraction system set at 40 °C and 15 MPa. After performing a static step for 10 min and a dynamic step for 20 min (flow rate of 2.0 L/min), an extraction yield of 85% could be obtained. Then the extract was deacidified, dehydrated and supplemented with carbonate solvents and lithium salts. Compared to commercial electrolyte, the recovered electrolyte exhibited acceptable electrochemical performance.

Though methods for electrolyte recovery including supercritical CO_2 and liquid CO_2 extraction have been developed in recent years, currently there is no large-scale applications. The difficulty lies in effective purification approaches, recovery of valuable lithium salts, and recycling cost.

5.3.2. Recycling of anode materials

Developing technologies to recover graphite from spent LIBs will further improve the economic feasibility of the recycling process, thereby enhancing the sustainability of the retired LIBs value chain.

Li et al. [149] proposed a graphite recycling method based on oxygen-free roasting and wet magnetic separation. The milled materials were roasted in nitrogen atmosphere for 30 min with a temperature of 1000 °C. The roasting products composed of Co, Li_2CO_3 and graphite. Then with the help of wet magnetic separation, Co will be magnetized and attached to the stirrer, Li_2CO_3 will dissolve, and graphite will precipitate and be collected by filtration. In laboratory scale experiments, the recovery rate of graphite could be 91.05%. Rothermel et al. [177] studied the feasibility of different graphite recycling methods. The flow-through subcritical CO_2 assisted electrolyte extraction method is preferred, because the recycled graphite shows the best cycling performance and the recovery rate of electrolyte is about 90%.

5.4. Policy and regulations

Government policies and regulations can undoubtedly help promote the reuse of EOL EV batteries. In fact, many countries and regions such as the United States, Japan, China, and the European Union, have released policies, regulations or subsidies to extend life and value of the spent LIBs [178].

The US has established a hierarchical framework for battery recycling at federal, state and local levels [11]. At the federal level, the Environmental Protection Agency (EPA) regulates the handling of hazardous batteries to protect human health and the environment. At the state level, most states follow regulations issued by Battery Council International (BCI) to guide retailers and consumers in battery recycling. At the local level, most cities have released recycling regulations to alleviate the hazards of spent batteries. China released the "Interim Measures for the Management of Recovery and Reutilization of Batteries of New-Energy Vehicle" in January 2018, to encourage the EV OEMs, battery manufacturers, and echelon utilization enterprises to treat second-life EV batteries in the principle of "echelon utilization and recycling priority". The measure clearly stated the liability requirements of relevant enterprises, which can promote innovation in reusing market mechanisms. In July of the same year, the Ministry of Industry and Information Technology (MIIT) of China issued "Interim Regulations on Traceability Management for Recycling of Power Battery of New Energy Vehicles", aiming to achieve the traceability of the EV batteries throughout their full life cycle [78]. Both regulations came into effect in August 2018.

5.5. Summary of recycling

EOL LIBs recycling can bring important economic and environmental benefits. While the development of recycling technology must deal with the different pack structure, diverse battery shapes and various active materials of the spent batteries. Although the recycling process is rather complicated, it can be roughly divided into two stages, namely pre-treatment and the valuable materials extraction.

During the pre-treatment process, the cells are removed from EV packs and then separated into valuable particles. The pre-treatment methods mainly include discharging, disassembly, crushing, screening and separation. Because cathode materials account for the highest proportion of value in spent batteries, the current recycling process is mainly focused on recovering high value metals from cathode materials and mainly include pyrometallurgical, hydrometallurgical, bio-hydrometallurgical and direct recycling technologies. In addition, recycling of plastics and graphite can further increase the value of the spent LIBs [146,179]. Combined recycling methods are performed to handle the problems of the high uncertainty of the composition of waste LIBs, in addition, online battery recycling system based on "Internet+" can help realize the recycling of spent batteries and effectively increase the recycling rate [180].

Besides the technical issues, national and local government policies, regulations and standards will also have significant impacts on the development of the LIBs recycling industry. Thanks to such regulations and standards, the battery recycling industry chain has made great progress in recent years.

6. Conclusion and future prospects

LIBs have been widely used for EV energy supply due to the merits such as high energy/power density, high reliability, and long life. The large-scale production and application of LIBs will inevitably lead to a large number of retired batteries and cause a shortage of raw materials. Due to potential economic, resource and environmental risks, the handling of retired LIBs must be comprehensively considered.

Remanufacturing, repurpose and recycling are sequentially arranged in the value chain. The ideal solution is to remanufacture the retired EV LIBs, then repurpose the cells, and finally obtain valuable materials by the recycling process. Remanufacturing, repurpose and recycling, as well as reduction and redesign, can form a "5R" strategy for retired battery management to obtain the maximum economic, environmental and resource value of LIBs. To fulfil the 5R principle, the following challenges and issues need to be considered:

i. Technical aspects: In the dangerous processes such as disassembly and pre-treatment, it is necessary to increase the automation degree, and the advanced technologies such as virtual operation, cloud-based methods, and CHAIN strategy, can be applied to avoid the hazards. Remanufacturing must overcome the challenges of safety assurance, evaluation method, screening and regroup technologies. Repurpose must deal with the complex management issues caused by changes in the dominant aging mechanisms. During recycling process, finding cost-effective and environmentally friendly recycling approaches with high generalization ability remains a huge challenge, and the recovery of anode materials and electrolyte may become a trend.

ii. Economic aspects: The economic benefits are the driving force behind the operation of CVC. Therefore, the cost of remanufactured and repurposed LIBs should continue to decrease to compete with the new batteries. Simple operating techniques, low-cost materials, reduced energy consumption, and environmental friendliness are required during the recycling process. In addition, Supply chains that benefit all participants should be established in the near future.

ii. Regulation and certification aspects: Undoubtedly, a more comprehensive regulation framework can effectively guide OEMs and related manufacturers to participate in battery CVC and promote the redesign of LIBs solution including process, structure and configuration. Customers can gradually build confidence in battery remanufacturing and repurpose with the help of certification and warranty.

iv. Data aspects: Information traceability and data security may also be critical in CVC process. Information about production, use, remanufacturing, repurpose and recycling should be fully interacted and shared throughout the lifespan. During this period, data security and data privacy issues must be considered. Advanced technologies such as big data, block chain and cloud-based technologies may be fully utilized to ensure the traceability and security of information.

Although reuse and recycling processes have great potential in dealing with the EOL LIBs, when faced with the massive amount of retired batteries, there are still some technical, economic, supply chain, and policy challenges that must be overcome. CVC process and 5R principle can provide a guidance for the large-scale industrial development of EV and energy storage applications.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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