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

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## Highlight

1. circular value chain based on 5R principle including reduce, redesign, remanufacturing, repurpose and recovery are proposed
2. key challenges of lithium-ion batteries remanufacturing and repurpose processes are analyzed comprehensively
3. up-to-date mainstream recycling approaches for electric vehicles lithium-ion batteries are investigated in detail
4. future prospects of lithium-ion batteries circular value chain are elaborated

## 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 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.

**Key words:** Lithium-ion battery, recycling, repurpose, remanufacturing, circular value chain

## **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], consequently the weight of spent EV batteries worldwide can weigh 1,700,000 tons by 2035.

The handling of end-of-life (EOL) LIBs must be taken carefully. 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]. 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 Ni may adversely affect environmental quality and human health. The lithium salts such as  $\text{LiPF}_6$  and  $\text{LiBF}_4$  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 with the help of various 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. Then the latest technologies to remanufacturing cycles are discussed in detail. A summary of improvement and challenges of repurpose process are presented from the view of 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 CVC are presented.

## **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.

EV packs usually consist 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]. But 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 cause the problem of high integration and assembly cost [18, 19]. Therefore, OEMs are tending to use prismatic or pouch designs to optimize system-level packaging efficiency. For instance, BMW i3 uses Samsung/SDI prismatic cells [7] and Nissan Leaf adopts pouch cells as the power supply [20]. The prismatic cell can improve space utilization and therefore have 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 good heat dissipation abilities [19]. However, the pouch cell is vulnerable and needs a special support structure to avoid swelling [22].

LIBs are composed of anode, cathode, electrolyte, separator collector and container [12]. The anode is usually made by graphite or lithium-titanate oxide (LTO); the cathode can be manufactured with lithium metal oxide comprised of  $\text{LiFePO}_4$  (LFP),  $\text{LiMn}_2\text{O}_4$  (LMO), lithium cobalt oxide (LCO), lithium-nickel manganese cobalt oxide ( $\text{LiNi}_x\text{Mn}_y\text{Co}_{1-x-y}\text{O}_2$ , 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  $\text{LiPF}_6$  and  $\text{LiBF}_4$ ) 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 and allows Li-ions to pass through [26, 27]. The collectors are usually produced by valuable metal such as aluminium (Al) and copper (Cu) [28, 29]; and Al is the primary material used in battery containers [30, 31].

## **2.2 Circular value chain of retired lithium-ion batteries**

Although different OEMs have adopted diverse solutions in EV application, the production processes of LIBs are very similar. Generally, the manufacturing processes can be roughly divided into multiple steps such as mixing, coating, calendaring, slitting, winding, welding and testing, etc. [32]. Then the LIBs are connected in parallel and series to form packs, which can be equipped in EVs as energy supply.

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 likely 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.

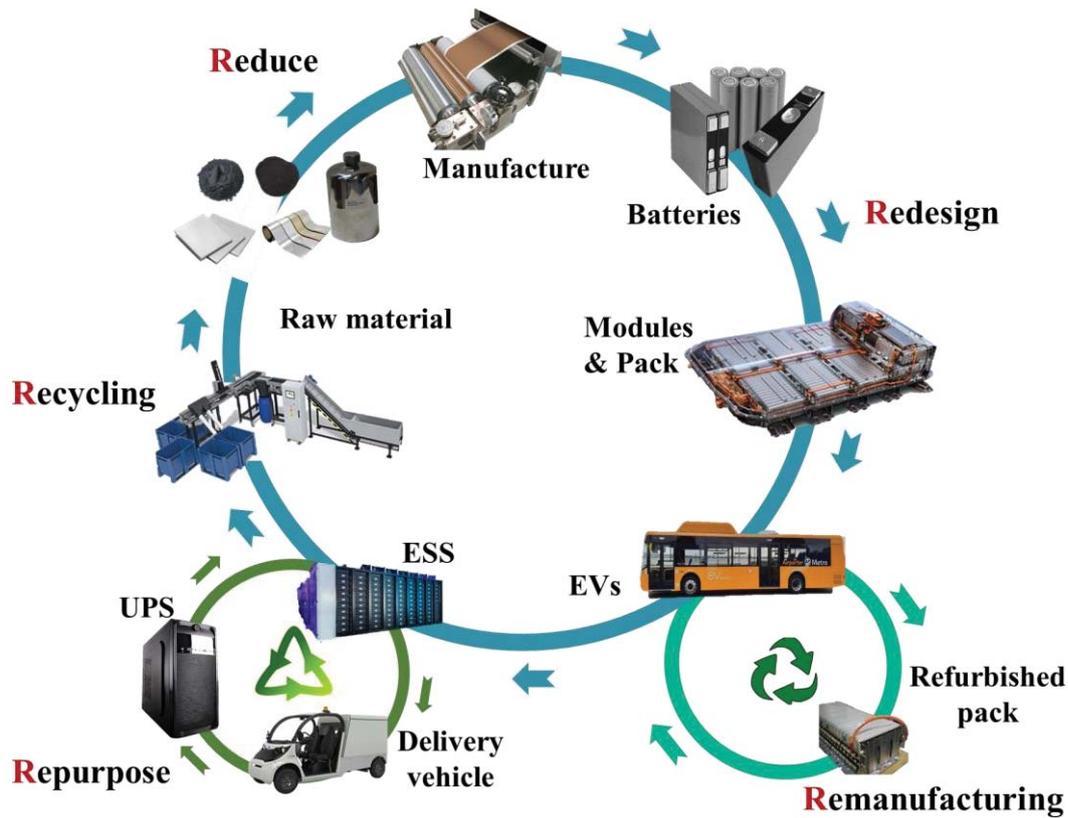


Fig. 1. Circular value chain of retired lithium-ion batteries [12, 24, 35]

With the help of evaluation technologies, the value of the retired EV LIBs can be optimized through the assessment result. 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, 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 aging, and those reused batteries will eventually be recycled after further degradation, thereby achieving a closed-loop CVC. In addition, redesigning the current pack configuration and structure 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 retired battery management to obtain the maximum economic, environmental and resource value of LIBs.

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 selection in EV LIBs processing [24], therefore not considered in the CVC process.

### **3 Remanufacturing**

After losing about 20%-30% initial capacity, LIBs can no longer satisfy the requirements of EVs such as driving range and acceleration [38]. While 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 how to deal with the retired LIBs. The final decision are made depending 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 result could be remanufactured in EV applications, and the schematic flowchart of the remanufacturing process is shown as Fig. 2.

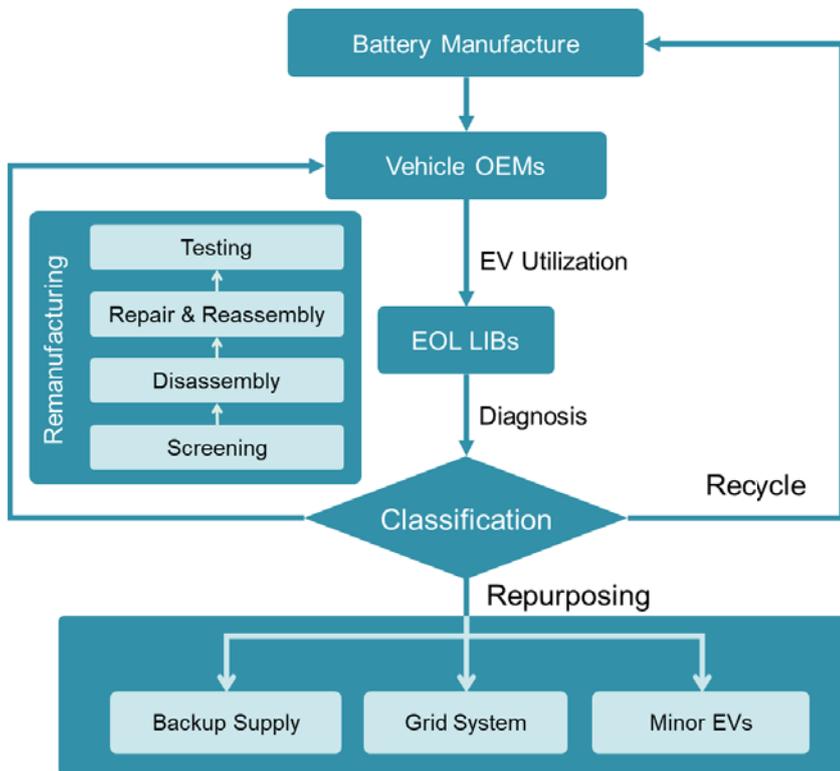


Fig. 2. Flowchart of the remanufacturing process

The remanufactured LIBs must meet all the requirements and standards specified by the OEMs, such as available capacity, power, and RUL. Due to cell-to-cell variations within the pack, LIBs usually degrade at different rates, and the decay of the EV pack is often caused by the failure of some special batteries, which may account for only a small part of the battery pack [15, 41]. Therefore, with the help of screening technology, the degraded batteries can be identified and replaced with new or qualified cells in other packs, thereby 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, partial pack disassembly, removal and replacement of degraded cells (repair), and reassembly of the pack [43]. Then the remanufactured packs can be applied for 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, thereby classify the destination of the second-life stages. 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 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.

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

Test	UL		IEC		SAE	IEEE	
	1642	2054	62133	62281	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	√						

However, such test methods 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.

Model-based and data-driven methods are developed to evaluate the safety characteristics of EV LIBs. Wang et al. [48] developed a safety performance model based on second-order oscillation feature to predict the failure displacement, and proposed a soft short circuit failure model to evaluate the safety performance. 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 safety operation range (safety envelope) 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 [50], and the relationship between abuse condition including voltage abuse and current abuse can be seen in Fig. 3. However, the practicability of SOS estimation methods needs further verification.

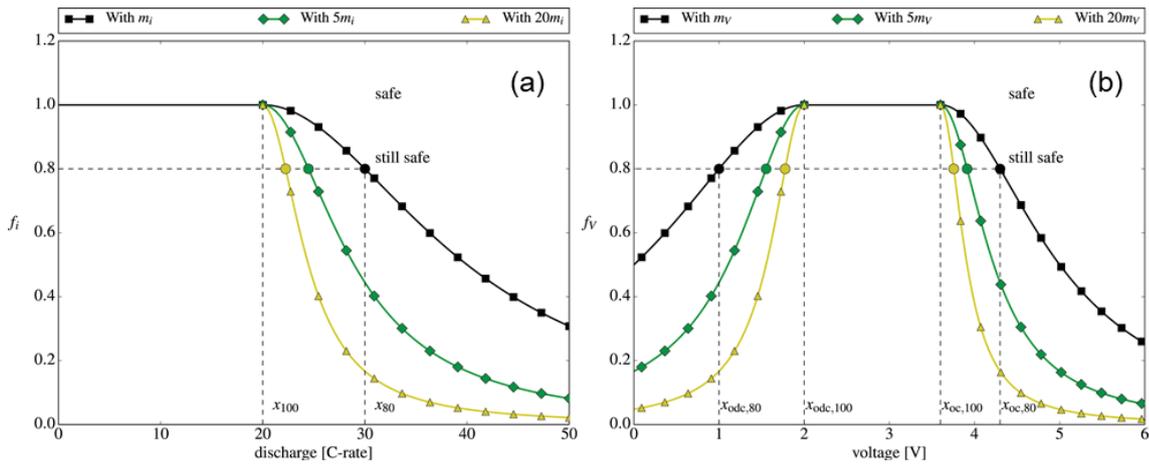


Fig. 3. SOS relationship with different abuse conditions [50]. (a) different current rate conditions. (b) voltage abuse including undervoltage and overvoltage.

### 3.1.2 SOH evaluation

SOH can quantify the degree of capacity decay and internal resistance increase, which may be caused from complex side reactions [45]. The majority of side reactions [32] is shown in Fig. 4 [51].

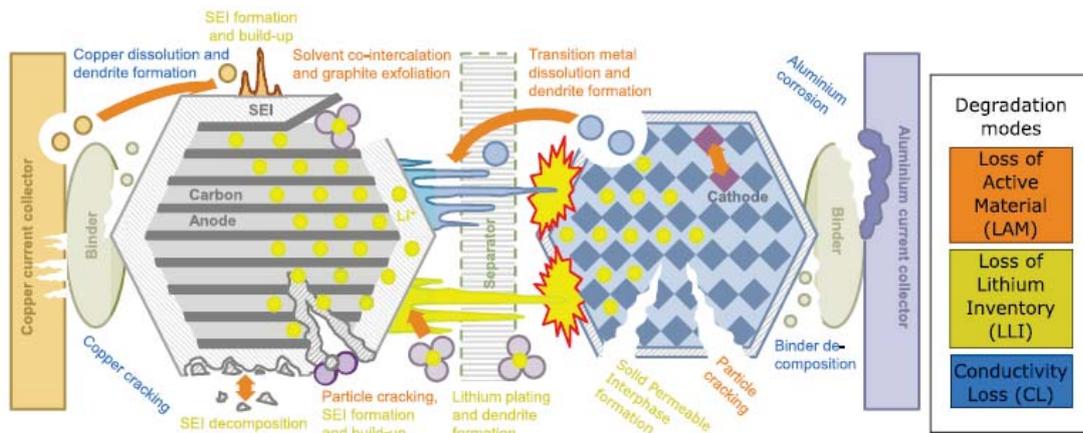


Fig. 4. Main degradation side reactions in LIBs [51]

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

Wang et al. [54] studied the capacity loss affected by different parameters and derived an

LFP 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 adaptability and accuracy are limited. Yang et al. [55] studied the aging behaviour of LIBs based on an electrochemical-thermal-mechanical coupled model, and researched the relationship between side reaction and working conditions. Experimental results show that the coupled method can be used to provide an accurate SOH prediction. However, high computational cost limits the applications of online application for 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 form the input layer. The NN model can estimate the SOH with low computation cost, and the training time of NN is about 21ms. Static and dynamic current profile tests prove the high accuracy of the NN-based method, where most of the SOH estimation errors are 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 can be obtained by calculating the grey correlation between the reference and comparative sequences. Experimental results show that the estimation error is limited to 4% and the average error is about 0.6%. Ladpli et al. [58] presented a SOH estimation method based on acoustic-ultrasonic guided waves. Preliminary experiments on pouch LIBs show that the information such as time of flight and signal amplitude will change during the aging process, and can be used for accurate SOH estimation through a statistical study.

Though numerous SOH estimation methods have been developed, most of them have not

been widely adopted in real applications until now. 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 maybe a possible direction for improvement.

### **3.1.3 RUL prediction**

RUL prediction is another critical method for retired LIBs evaluation, which is a subjective 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 decays during both storage and operation process, corresponding to calendar life and cycling life respectively. 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, ECM models, EMs, artificial intelligence (AI) methods, and hybrid methods, can also be adopted for RUL prediction [60, 61].

Lyu et al. [62] proposed a novel EM-based PF framework to predict RUL, in which state variables were selected from internal SOH-related features. The methodology combined the electrochemical mechanism of the battery with data-driven PF method, and can achieve high quality and high robust RUL prediction applications for different types of batteries. When the beginning of prediction (BOP) is 50 cycles, the RUL prediction error is about 15.5%, and when the BOP is 80 cycles, the error is less than 5%. The reason is that more historical information can bring more accurate prediction results. The main challenge of the EM-based RUL 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 the residual energy

(remaining life) for retired EV batteries, in which genetic programming approach was utilized to predict the remaining capacity as a function of various applied stresses. 1510 groups of experimental data are obtained to verify the feasibility of the approach. Zhang et al. [64] developed a RUL prediction method with the help of random forest (RF) and recursive least square (RLS) algorithm. The RF algorithm is used to identify the battery aging stage, and the RLS algorithm with forgetting factor is adopted to predict the EOL cycle based on aging stage. The calculation can be completed within 0.3 s, and when the BOP is greater than 300 cycles, most of the prediction errors are less than 10 cycles. The experimental results show 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 be replaced with qualified cells in following steps. The screening process can be performed based on differences in parameters or properties, or based on data-driven methods.

Liao et al. [65] believed 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 demonstrate the correlation between Warburg impedance, charge transfer resistance and LIB capacity, which can be weighted by Pearson's correlation coefficient. And 20 batteries in good conditions are 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 and concluded that the variations of Li

inventories is the dominant capacity aging factor. Quantitative analysis results show that the changes in IC peaks and peak areas can represent the variations of anode materials, lithium inventories and reaction kinetics, therefore ICA result can serve as the basis for high-precision screening strategies.

### **3.3 Disassembly, reassembly and test**

Disassembly 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]. But until now there is no standardization for the configurations and structures of 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 disassembly steps [70]. Another advanced technology for battery disassembly is cloud-based method, 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 can be reduced by more than 50%.

The reassembly process is very similar to disassembly steps, and because the remanufactured packs are used for the same purpose, the original test methods are still

applicable. In addition, the current 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 the cost of about \$2,850 (24 kWh) [74].

Though remanufacturing can save about 40% 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 the ones 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 is another challenge that the large-scale industry applications have to face [76].

## **4 Repurpose**

Repurpose is somewhat similar to remanufacturing, especially for evaluation methods,

screening methods, and disassembly processes. However, because the repurposed LIBs will begin their second life in different scenarios, the pack structure may change, new software and/or hardware for BMS may be applied [77], and different TMS and EMS can be used. 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 overcharging, over discharging, accelerated aging, and even thermal runaway (TR) may increase [6]. Therefore, it is essential to regroup the batteries with similar characteristics such as capacity, internal resistance and open circuit voltage (OCV).

Multiple algorithms can be utilized for LIBs regroup, including parameter-based method and dynamic method [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 show that compared with the traditional k-means method, the improved method has similar performance and can reduce the convergence time by about 79%. Xu et al. [6] proposed a clustering method for retired LIBs regrouping based on traversal optimization, which can form clusters with high differences between clusters and low variations within clusters. Experimental results show that the proposed regrouping method are more effective than k-means algorithm and affinity propagation (AP) method, and are applicable for

large samples applications. Zhou et al. [80] proposed a rapid classification method for retired EV LIBs, which can regroup the batteries depending on capacity and internal resistance with the help of piecewise linear fitting (PLF) and radial basis function neural network (RBFNN) methods. Data set can be collected by PLF method, then trained by RBFNN for large-scale LIBs classification. 4 Cells with similar characteristics are selected from 12 retired batteries and connected in series to form a pack. At the end of discharge, the voltage difference of the pack is about 34 mV, and the root mean square error (RMSE) is about 15.5 mV. Experimental results prove that the proposed regroup method can 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 popular adopted due to the merits of simple structure and good scalability, but the parameters need to be adjusted according to working 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 real 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 by 81%. Yang et al. [86] proposed a multi-time scale EKF method for SOC estimation of aged battery pack. With the help of special and difference model based on second order ECM, the SOC of the special battery, remaining cells, and the pack, can be estimated in different time scales. Under New European Driving Cycle (NEDC) and Urban Dynamometer Driving Schedule (UDDS) dynamic conditions, the maximum error of SOC estimation is about 4.6% and 3.9% respectively, which proves 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 roughly divided into characteristic map (CM)-based methods and model-based methods [81]. CM-based methods can estimate SOP based 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 show that within the range of 10% to 95% SOC, the peak discharge current error of the JEVS method is less than 10%, which proves the accuracy of the improved method. Wei et al. [88] developed a novel parameter identification (PI) method based on first order ECM with hysteresis, which can obtain parameters online with the help of extreme seeking algorithm and obtain SOP estimation results considering the voltage and current limitations. The simulation results on real data prove the feasibility and stability of the proposed PI and SOP estimation methods. Zhou et al. [89] proposed a EKF-based low computational cost SOP estimation method for series-connected batteries, which can select the representative battery depending on the variation of characteristic voltages and ohmic resistances. The experimental results under dynamic condition at different temperatures prove the accuracy, feasibility and robustness of the proposed estimation method.

### **4.2.3 SOH Estimation Methods**

Accurate SOH estimation, which can help realize 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 face the challenges of computational cost and PI.

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 can be used to identify the battery SOH with the help of charging data. Test results prove the feasibility and generalization of the SOH estimation method, which has the potential to be utilized in real BMS. Zhang et al. [91] discussed the capacity performance of 24 retired LFP modules using 6 different capacity test protocols, and concluded that the constant charge of 1/3 C can strike a balance 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 (ARMSE).

#### **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 good 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 depending on the improved RLS algorithm. With the updated capacity from SOH estimation, the accuracy of SOC estimation can be significantly improved. Under dynamic stress test (DST) and Federal Urban Driving Schedule (FUDS) dynamic conditions, the estimation errors of SOH and SOC are 0.5% and 1.1% respectively, which prove the effectiveness and accuracy of the proposed

method. Hu et al. [94] developed a hierarchical framework for SOC, SOH and SOP co-estimation on multiple time scales. The framework includes 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 verify the feasibility of the proposed method with a SOC estimation error less than 3%, a voltage error within 25 mv, and a capacity error less than 3% respectively. Yang et al. [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 temperature sensitive and usually achieve the best performance near room temperature. TMS can help maintain the maximum temperature and temperature difference in 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, and 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 can help maintain the pack temperature in optimum range for several days. The simulation result show that when the heating plate is 0.16 m<sup>2</sup> and placed horizontally under the pack, the maximum temperature difference between the batteries is less than 5 °C. Compared with other size and arrangement, the temperature uniformity performance are improved. White et al. [99] compared the performance of various retired EV LIBs in grid frequency regulation (FR) applications. Experimental results show that the thermal characteristics of second life batteries are greatly affected by TMS and working conditions. Under aggressive FR conditions, the peak temperature of Lishen and Leaf batteries (with passive TMS) reach 48.7 °C and 49.6 °C respectively, while the peak temperature of batteries with active TMS (such as Tesla and Volt) is below 40 °C. In addition, active TMS can achieve better heat accumulation and temperature fluctuation performance, thereby are 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 time of charge-discharge cycles in EV applications, packs will suffer from inconsistencies in capacity and internal resistance. Such imbalance may exacerbate and lead to high risks of system failure and safety issues during reuse period. Fortunately, EMS can improve imbalances, 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 evaluation result of inconsistencies of capacity and internal resistance between the retired batteries, equalization approach based on the capacity utilization was adopted 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 show that the equalization circuit can 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 has a balancing ability up to 1.08 A and can fulfil the equalization requirement of 320 Ah batteries with significant imbalance. The off-grid ESS with high reliability has been applied on an island in Tanzania.

## 4.5 Applications of repurpose

Repurposed batteries can be utilized in less-stressful 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.

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	4MWh/ 4MW
 Backup supply for telecom tower in China [78]	Stationary	China Tower	30kWh
 EV Charger in California [106]	Quasi-stationary	Siemens, Freewire	80kWh/ 11kW
 Delivery vehicles in China [78]	mobile	SF Express	2-3 kWh

### 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 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 to reuse 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 real data show that the solution can 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 can congregate grid-able EVs and repurposed batteries together to serve as backup systems for the grid. Simulation results showed that such model can save 70% of the cost compared with conventional solutions, and can 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 structure, diverse battery shapes and various active materials [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. The majority of anode materials uses graphite, 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, the majority cathode materials are LFP and NMC family [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 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 from cathode materials of spent LIBs [120]. Although the recycling process is very complicated, it can be roughly divided into two stages. The first stage is pre-treatment, includes discharging, disassembly, crushing, screening and separation, and the purpose of which is to remove cells from EV packs and separate them into valuable particles that can be used in the next process [7, 28, 121, 122]. Materials extraction is the second process, 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 purpose of pre-treatment is to separate and enrich valuable components and materials from EOL LIBs based on differences in physical properties (such as shape, density and magnetic properties). Pre-treatment can help increase recovery rate, reduce energy consumption, avoid the safety risks and reduce the 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 could be roughly divided into discharging, disassembly, crushing, and separation processes, which is shown in Fig. 5 [122, 128].

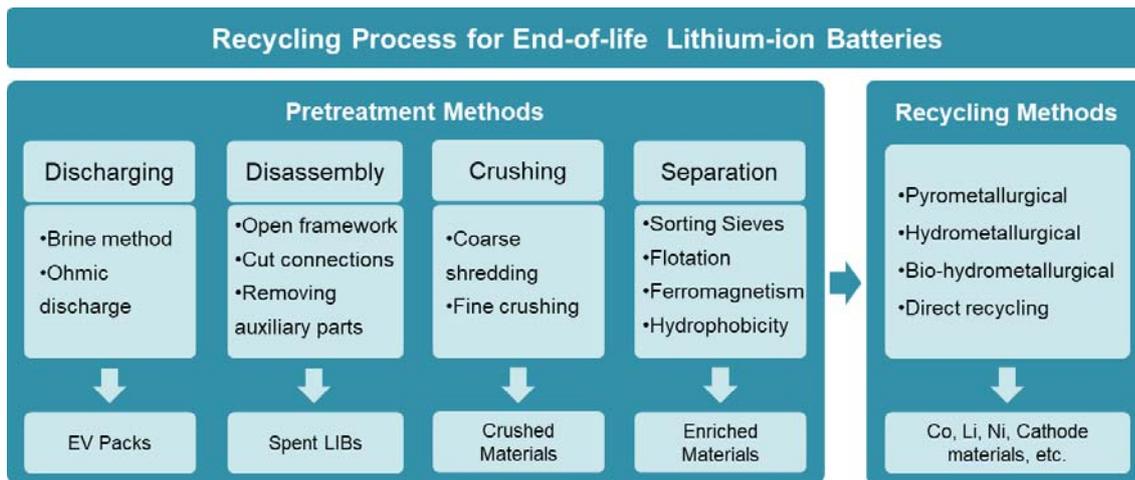


Fig. 5. Main procedure of EV LIBs pre-treatment [122, 128]

### 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 refers to soak the used LIBs in solution such as NaCl and Na<sub>2</sub>SO<sub>4</sub> for a sufficient time to completely discharge. Nie et al. [131] immersed the LCO batteries in Na<sub>2</sub>SO<sub>4</sub> saturated solution for 24 hours 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, otherwise high rate of electrolysis may occur and release vigorous gases [7].

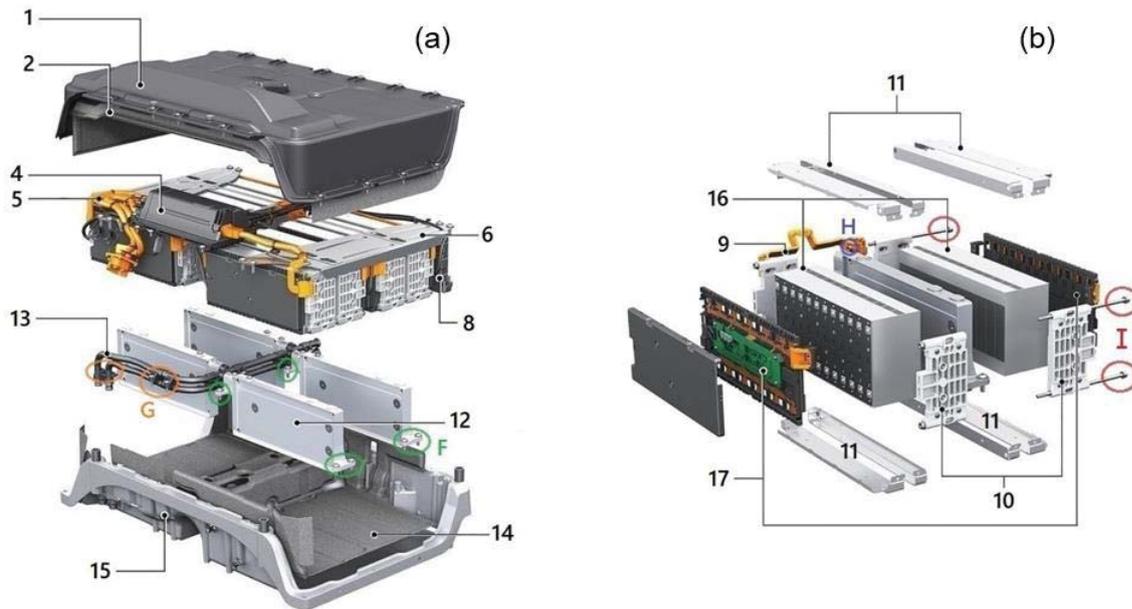
The Ohmic discharge method refers to discharge the battery with an external load circuit. Kruger et al. [132] discharged the NMC batteries with resistances and then recycle the active materials. He et al. [125] discharged 24 series-connected cells by connecting to the load, and the batteries can be discharged to 2 V synchronously within 2 hours. During the process, a BMS

with temperature sensors was used to monitor the pack status. Ohmic method can fully discharge the spent LIBs in a relative short time. But a large amount of heat can accumulate in a short time, and high temperature may cause the risk of TR.

### **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 pre-treatment process, the battery system can 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 purpose of disassembly process is to minimize environmental costs and maximize economic benefits [133], and many advanced technologies are used for LIB disassembly. As described above, robotic assistance, virtual disassembly, and cloud-based technique may be the tendency of disassembly technologies.



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

Fig. 6. Exploded view of the battery system and the module [27]

### 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 safety 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 show that the pieces with size greater than 6 mm are mostly

casings and separators, and the graphite anode and the cathode active materials form 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 seconds, 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 demonstrate that the coarse products are mainly formed by metallic shell, collector foils and organic materials, and the fine products mainly contains graphite and the cathode particles, which can be recycled in the subsequent metallurgical process.

#### **5.1.4 Separation**

After shredding and crushing process, the various materials are procedurally separated and conditioned for further treatment steps [134]. Physical processes can be used to separate and enrich of 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) is mainly composed of the active materials from the electrode, accounting for about 19.5% of the total weight. The proposed separation process can effectively separate materials, thereby improving the purity of the products, and has the potential for large-scale application. Yu et al. [138] proposed a flotation-based separation process to separate graphite and LCO materials from spent LIBs. The obvious wettability difference between the electrode materials contributes to a concentrate grade of 97.13% and 73.56% respectively.

Grinding flotation has the characteristics of high efficiency and pollution free, which is helpful for 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.

### **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 TR or even explosion. The disassembly step still needs human participation due to the diverse structure of 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 may have the potential for large-scale applications.

## **5.2 Metallurgical Technologies**

Various materials such as Co, Li, nickel (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 may be the most popular technology for 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 process various unstable materials [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 roughly 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 and 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 with 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 hour and at 800 °C for 2 hours separately. Then the powder was mixed with supplementary  $\text{Li}_2\text{CO}_3$  and calcined at 850-950 °C for 12 hours to obtain  $\text{LiCoO}_2$ . The proposed method has the advantages of simple process and environmental friendliness, and the physical and electrochemical properties of the  $\text{LiCoO}_2$  products can meet the requirements for commercial application. Li et al. [149] proposed a pyrometallurgy method to recycle valuable materials in situ from electrode materials including  $\text{LiCoO}_2$  and graphite. The mixture of Co,  $\text{Li}_2\text{CO}_3$  and graphite can be obtained by calcining at 1000°C for 30 minutes, and the recovery rate of Co, Li and graphite can 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 can 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

minutes. The mixed electrode materials including graphite and LMO were heated at 800°C for 45 minutes, and the optimal recovered rate of Li is 91.3%, the purity rate of Mn<sub>3</sub>O<sub>4</sub> was 95.11%, and the purity rate of Li<sub>2</sub>CO<sub>3</sub> 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<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub>, 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<sub>2</sub>O<sub>2</sub> 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 removal of 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  $\text{H}_2\text{O}_2$  as additional reductant can leach more than 96% Co and almost 100% Li, and had better performance in terms of efficiency and environmental friendliness than HCl and  $\text{H}_2\text{SO}_4$ . 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 minutes, and then transition metals (Ni, Co and Mn) enriched residues were completely leached in sulfuric acidic medium at 70 °C for 30 minutes. Next, the transition metals can be sufficiently precipitated by  $\text{Na}_2\text{CO}_3$  and  $\text{NH}_3\cdot\text{H}_2\text{O}$  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 condition of oxalic acid concentration of 0.3 mol/L, solid/liquid ratio of 60 g/L, reaction time of 60 minutes, and reaction temperature of 80 °C, the proposed method can extract about 98% of Li and 92% of Fe in terms of  $\text{FeC}_2\text{O}_4\cdot 2\text{H}_2\text{O}$ .

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. Therefore, 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 sulphate leaching reagents. Under the optimum conditions at 80 °C for 5 hours, 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  $\text{NH}_3 \cdot \text{H}_2\text{O}$  and  $\text{NH}_4\text{HCO}_3$  is 120 g/L and 75 g/L respectively at 80°C for 4 hours), the process yields 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 can effectively accelerate the selective leaching reaction. When using  $\text{NH}_3\text{-NH}_4\text{Cl}$  solutions mixed with  $(\text{NH}_4)_2\text{SO}_3$  as a reductant, leaching process can extract 100% Co, 98.3% Ni and 90.3% Li. And the proposed method can 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, which 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 show that the *A.n* bioleaching method can dissolve 82% Co and 100% Li under the optimal conditions, which is more effective than the *A.t* bioleaching or acid leaching, and has 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 is about 99.2% Li, 50.4% Co and 89.4% Ni. Experimental results prove the effectiveness and environmental harmless 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 handling 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 the first treatment, the recovered LFP materials were further directly regenerated with  $\text{Li}_2\text{CO}_3$  at 600-650 °C for 1 hour under Ar/ $\text{H}_2$  gas flow. The products perform 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. The experimental results demonstrate the low cost and feasibility of the direct regeneration process. Shi et al. [169] proposed a green energy-saving method for the regeneration of LCO cathode materials, which can 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 can also handle batteries with different degradation status by eliminate the compositional and structural defects[170]. Experimental results show the potential to be used for large-scale recovery of EOL LIBs cathodes.

### **5.2.5 Comparison of Recycling 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 research focus for EOL LIBs recycling. However, this recovery methods have to face 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 require long time cultivation and are 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.

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

Although different technologies have been researched to recover valuable metals from used LIBs, the overall recycling performance such as economy, environment, and timeliness are still unsatisfactory due to the high uncertainty of the composition of waste LIB waste. Therefore, combined recycling processes can be performed [4, 171]. For instance, Sun et al. [172] proposed a novel process combining vacuum pyrolysis 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  $\text{LiCoO}_2$  and  $\text{Ni(OH)}_2$ .

## 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 easy purification process [174].

Grützke et al. [175] compared the extraction behavior between supercritical  $\text{CO}_2$  and liquid  $\text{CO}_2$  methods. Supercritical  $\text{CO}_2$  can achieve higher recovery rate of cyclic carbonate such as EC, while Liquid  $\text{CO}_2$  can extract higher rate of linear carbonates and achieve higher overall recovery rate of carbonates. Additional solvents can further improve the extraction of electrolytes. The best electrolyte overall recovery rate for a Panasonic 18650 cell (NMC/graphite) can reach  $(89.1 \pm 3.4)$  wt%. Mönnighoff et al. [176] performed the supercritical  $\text{CO}_2$  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 minutes, and then a  $\text{CO}_2$  flow-through step with addition of acetonitrile for 5 minutes. After repeating the procedure for several times, the extraction of electrolyte was completed. Liu et al. [174] developed a supercritical  $\text{CO}_2$  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  $\text{CO}_2$  extraction system set at 40 °C and 15 MPa. After performing a static step for 10 minutes and a dynamic step for 20 minutes (flow rate of 2.0 L/min), an extraction yield of 85% can be obtained. Then the extract was deacidified, dehydrated and supplemented with carbonate solvents and lithium salts.

Compared to commercial electrolyte, the recovered electrolyte exhibits acceptable electrochemical performance.

Though methods for electrolyte recovery including supercritical CO<sub>2</sub> and liquid CO<sub>2</sub> 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 minutes with a temperature of 1000 °C. The roasting products composed of Co, Li<sub>2</sub>CO<sub>3</sub> and graphite. Then with the help of wet magnetic separation, Co will be magnetized and attached to the stirrer, Li<sub>2</sub>CO<sub>3</sub> 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<sub>2</sub> 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 utilization of New energy vehicle power battery” 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 of the 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 face 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 LIB waste, 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. However, 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 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 face the challenges of safety assurance, evaluation method, screening and regroup technologies, while 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 be 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, and 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.

- iii. 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.

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 faced. CVC process and 5R principle can provide a guidance for the large-scale industrial development of EV and energy storage applications.

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