Research on Power Battery Aging and Green Recycling Technology

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ABSTRACT

With the acceleration of global carbon neutrality, the new energy automobile industry has experienced explosive growth, which has brought more and more pressure on resources and environment due to the wave of power batteries that are about to be retired. Accurately describing the degradation path of batteries and designing economical and feasible green recycling solutions have become the main concerns of academia and industry. Therefore, this paper proposes a narrative review, covering four aspects: the explanation of aging mechanism, the prediction of health status (SOH), the evaluation of recycling technology, and the tracking of carbon footprint through digital twins. On the mechanism level, we synthesize the clutch framework of lithium inventory loss (LLI) and movable material loss (LAM) to explain the multi-scale failure modes, including the pairing scheme of cathode stripping, anode SEI evolution and electrolyte decomposition. At the prediction level, we compare and analyze the advantages of equivalent circuit model, semi-empirical degradation model and machine learning method, and emphasize the potential of mechanical data fusion channel engineering application. In terms of recycling, we quantitatively compare Pyro Metallurgic, Hydrometallurgic and direct recycling methods in terms of metal recovery, energy consumption and carbon emissions. This reveals the great advantages of non-destructive recycling technology in reducing carbon emissions. In terms of traceability, we introduced the concept of digital twins and put forward a preliminary framework for tracking carbon footprint. The framework covers the whole supply chain from design and operation to recycling. The results provide a theoretical reference for optimizing the decision-making in the battery recycling system.

Keywords: Power Battery; Aging Mechanism; SOH Prediction; Green Recycling; Digital Twin.

1. INTRODUCTION

The establishment of carbon footprint and carbon neutrality goal has promoted the new energy automobile industry to embark on a very rapid development path. According to the data of China Automobile Manufacturers Association, by 2025, the number of new energy vehicles in China will exceed 35 million, and the resulting waste batteries are expected to reach 820,000 tons. By 2030, this figure may reach 3.5 million tons. Such a large number of waste batteries pose an unprecedented challenge to resource recovery and environmental protection. Although the Ministry of Industry and Information Technology issued the Management Measures for Cascading Use of New Energy Vehicle Batteries, which clarified how to build a recycling system, the complexity of aging mechanism, the economy of recycling process and the quantitative assessment of environmental impact are still the main obstacles restricting the healthy development of the industry.

The existing research usually regards battery aging and recycling technology as two independent topics, but pays little attention to the deep relationship between them. In fact, the aging mode of the battery-mainly the loss of lithium (LLI) or the loss of active substances (LAM)-directly affects the selection and optimization of the subsequent recovery process. For example, batteries aged mainly due to LLI have a relatively complete cathode material structure, which makes them more suitable for direct repair and regeneration technology. For the batteries dominated by LAM, the material structure is seriously damaged, and thermal metallurgy or hydrometallurgy may be a more practical choice. This vision of linking aging and recycling is the core idea that this paper tries to establish.

Based on the above reasoning, this paper systematically investigates this field from four dimensions: firstly, the multi-scale aging mechanism of power batteries is analyzed, and the relationship between micro-material failure and macro-performance degradation is clarified; Secondly, compare the applicable scenarios and accuracy limitations of different SOH prediction methods; Third, quantitatively evaluate the resource efficiency and environmental load of the main recycling technology routes; Fourthly, the digital twin framework is introduced to explore the technical feasibility of carbon footprint traceability throughout the life cycle. Through this analysis, our goal is to provide a theoretical basis for building a closed-loop recycling system of batteries.

2. MULTI-SCALE AGING MECHANISM ANALYSIS OF POWER BATTERIES

2.1. Correlation Between Macroscopic Electrochemical Characteristics and Microscopic Material Evolution

The aging of power batteries is not caused by one thing, but by several forces acting together, such as electricity, heat and mechanical force. On the macro level, capacity loss and impedance increase are the two most easily seen signs of aging. But behind these signs, there are complex mechanisms at the material level. After years of experiments tracking Coulomb efficiency, Dahn team [1] found that in the early cycle, a very small amount of active lithium would be irreversibly lost, and this loss would increase exponentially with the increase of cycle depth. Ouyang Minggo team [2] used an experimental platform with various conditions to show the relationship between lithium deposition on the anode and the risk of thermal exposure. They pointed out that rapid charging at low temperature is the key external stress factor leading to abnormal lithium electroplating.

Through advanced technologies, such as neutron diffraction and in-situ transmission electron microscopy, researchers can observe how the materials of batteries change at the atomic level. It has been proved that the microcrack diffusion in the high ternary cathode (NCM811) is directly related to the anisotropic shrinkage along the network C axis during charging and discharging [7]. These microcracks not only increase the impedance of charge transmission, but also create a path for electrolyte penetration. This aggravates the secondary reaction to the interface and forms a vicious circle. At the same time, the SEI film on the surface of graphite anode is not static-it breaks and reconstructs again and again in the continuous dissolution/dissolution process. Use a little more active lithium and electrolyte for each reconstruction.

2.2. Decoupling Analysis of Typical Aging Modes

In order to quantitatively distinguish the relative contributions of different aging mechanisms, the LLI/LAM separation framework proposed by Vetter et al. [3] is widely used. The main idea of this framework is to decompose the capacity loss of the whole battery into three independent parts: cathode capacity loss, anode capacity loss and active lithium loss. Incremental capacity analysis (ICA) and differential voltage analysis (DVA) are used. However, in practical application, this method encounters problems such as the demand for high-quality data and high dependence on operating conditions. Peterson et al. [5] show that the accuracy of identifying ICA characteristic peaks is significantly reduced under dynamic operating conditions, which limits the direct use of this method in embedded battery management system (BMS).

In recent years, the “reservoir” model, as an extension of the LLI /LAM framework, has brought a new method to understand how the aging mechanism changes with time. The model assumes that there is some “hidden” capacity losses-they are lithium ions, because they are trapped by secondary reaction products, but they can resume participating in electrochemical reactions under some conditions. The introduction of this concept explains the phenomenon of capacity “rebound” seen in some experiments, and increases the uncertainty when trying to evaluate the capacity usage in cascade scenarios. Generally speaking, the accurate identification of aging mechanism is still an unresolved problem, which requires the common progress of characterization technology and data analysis methods.

3. STATE-OF-HEALTH PREDICTION: FROM MECHANISM MODELS TO DATA FUSION

3.1. Advantages and Limitations of Mechanism-Driven Methods

Equivalent circuit models (ECM) are widely used for real-time battery state estimation in management systems, because they have fast calculation speed and their parameters have physical significance. First-order and second-order RC models can meet different accuracy requirements. But their common limitation is that their parameters change with battery aging. If there is no regular online identification, the estimation error of SOH will increase rapidly. Pseudo-two-dimensional (P2D) porous electrode model [14] can describe the transport of lithium ions in solid and liquid phases from the basic principle. But to solve this model, a lot of computing power is needed. This is why it is mainly used for off-line simulation and optimization design.

The semi-empirical degradation model attempts to be both physically understandable and fast to calculate. Arrhenius equation is widely used to describe the influence of temperature on aging speed, and the power law relationship between cycle times and capacity loss has been confirmed by a large number of experimental data. The main challenge of these models is that their coefficients depend largely on the specific battery chemistry and usage conditions, and they do not work well in all brands and models. As Birkl and Howey [4] said, “There is no general degradation model, only an approximation of a specific application scenario.

3.2. Data-Driven and Fusion Methods

Machine learning method opens up a new way to predict SOH [15]. Support Vector Regression (SVR), Gaussian Process Regression (GPR) and the new deep learning model demonstrate the powerful ability to deal with complex nonlinear relationships. Shen et al. [6] used convolutional neural network (CNN) to extract the aging signal directly from the charge-discharge curve, and successfully predicted the error of SOH under laboratory conditions is less than 2%. However, the problem of data-driven method is also obvious: training data must cover many use conditions and aging stages. Otherwise, when the models encounter too many different data, they may produce very big prediction errors.

The fusion path combining mechanical model with data-driven method is considered as the best solution to balance the accuracy of prediction and generalization ability. A common strategy is to use a mechanical model to generate prior knowledge, and then use a data-driven model to correct the residue. For example, circuit model equivalence is first used to calculate the basic SOC and SOH estimates, and then the neural network to learn the nonlinear deviation not captured by the model. The other method integrates physical constraints into the loss function to ensure that the output of neural network conforms to basic physical laws, such as mass conservation and energy conservation. These physical information neural networks (PINN) are still in the exploratory stage in the field of batteries, but they initially show the potential to improve the extrapolation ability.

4. COMPARISON AND EVALUATION OF GREEN RECYCLING TECHNOLOGY ROUTES

4.1. Current Status and Bottlenecks of Pyrometallurgical and Hydrometallurgical Processes

As the first commercial battery recycling technology, pyrometallurgy has been widely used in early recycling centers in Europe and the United States because its process is simple and it can handle a large number of batteries [8,9]. This process converts the metal oxides in the battery into alloys at very high temperature, but lithium easily escapes into waste or smoke due to heat, and the recovery rate is usually less than 50%. Considering the strategic importance of lithium resources, such a low recovery rate cannot meet the needs of sustainable development. In addition, pyrometallurgy consumes a lot of energy: 500-800 kWh per ton of battery, which produces more carbon than other recycling methods.

Aluminum hydride is the main technology of battery recycling in China at present. Companies such as GEM and Huayou Cobalt already have large factories [11]. The method uses acid or alkali to dissolve precious metals from cathode materials. Then, these metals are separated and purified by extraction, precipitation and electrospinning, and finally the metal salt with battery quality is obtained. Under proper conditions, more than 98% of lithium and more than 85% of lithium can be recovered. But hydrometallurgy has a cost to the environment. It uses many acidic or alkaline chemicals and produces wastewater containing heavy metals and acidic gases. The construction and operation costs of these waste treatment plants are very high, accounting for a large part of the total cost.

4.2. Direct Repair and Regeneration Technology

Different from the “destructive reconstruction” methods of pyrometallurgical and hydrometallurgical methods, direct repair technology tries to restore electrochemical properties while retaining the main structure of materials. The theoretical basis of this concept is that the latex structure of the cathode material is basically not destroyed for the old batteries mainly with LLI, and it can only be reused by replacing the lost lithium and repairing the surface defects. Recently, researchers reported a high-temperature impact regeneration strategy, which can complete lithium replenishment and structural repair of waste $LiFePO_4$ cathode in tens of seconds [10]. The electrochemical properties of recycled materials are close to the original level. Compared with traditional hydrometallurgical recovery, this method can reduce energy consumption by about 70%, and at the same time avoid producing acidic and alkaline liquid waste, showing significant environmental benefits.

However, the industrialization of direct repair technology still faces some difficulties. First of all, this method requires very uniform raw materials. If batteries with different aging degrees or batteries with different chemical systems are mixed, the maintenance efficiency is much lower. Secondly, to correctly adjust the process parameters, we must first accurately understand how the battery is aging. However, the current aging diagnosis technology is not enough to classify a large number of batteries. Thirdly, for the products repaired by this direct method, there is still a lack of common rules and enough tests to verify whether they are all as effective as each other and last for a long time. In order to solve these problems, experts from different fields need to work together: material science, electrochemical engineering and automatic sorting technology.

Table 1. Comprehensive Comparison of Three Recycling Technology Routes.

Evaluation Index	Pyrometallurgy	Hydrometallurgy	Direct Repair
Li Recovery Rate	<50%	>85%	N/A
Co/Ni Recovery Rate	>95%	>98%	N/A
Energy Consumption (kWh/ton)	500-800	300-500	80-150
Carbon Emissions (kg CO2/ton)	1200-1800	600-1000	150-300
Process Maturity	Industrialized	Industrialized	Lab/Pilot
Environmental Load	High	Medium-High	Low
Regenerated Material Performance	Requires reprocessing	Requires resynthesis	Near-native

Note: Data derived from reported experimental and techno-economic analysis results in the literature.

As shown in Table 1, the three recycling technology paths show great differences in resource efficiency, energy consumption and environmental impact.

4.3. Construction of Multi-Dimensional Evaluation Index System

In order to systematically compare all the advantages of different recycling technology routes, this paper constructs a three-dimensional evaluation matrix. It includes resource efficiency, environmental impact and economic feasibility. The dimension of resource efficiency includes the recovery rate of major metals, such as lithium, aluminum chloride and metals, and the performance indicators that renewable materials can achieve. The dimensions of environmental impact include energy consumption per unit volume of treatment, greenhouse gas emissions and the generation of wastewater and waste gas. The dimensions of economic feasibility include capital investment intensity, operating cost and added value of products. Based on reported experimental data and techno-economic analysis in the literature, hydrometallurgy performs optimally in resource efficiency, direct repair shows significant advantages in environmental impact, while pyrometallurgy only maintains leadership in processing scale and process maturity.

5. FULL LIFE CYCLE DIGITAL TWIN AND CARBON FOOTPRINT TRACEABILITY

The EU “New Battery Regulation” effective in 2023 [12] imposes strict sustainability requirements on power batteries, including carbon footprint declarations, minimum recycled material content ratios, and innovative systems such as the battery passport. Among these, the “Battery Passport” requires recording key information throughout the battery’s full life cycle and making it accessible to supply chain parties and consumers through carriers such as QR codes. The implementation of this system objectively requires a digital infrastructure covering the entire chain from design, manufacturing, operation to recycling, for which digital twin technology provides a feasible technical pathway [13].

The core concept of digital twins is to create a high-precision virtual mirror image of physical objects, and monitor, predict and optimize the synchronized physical process through real-time data interaction. In the field of power battery, digital twins can be divided into five functions: physical layer (physical structure of battery, module and battery pack and sensor network), data layer (collecting and cleaning heterogeneous data, such as BMS operation data, maintenance records and environmental parameters) and model layer (model based on mechanism or data, such as electrochemical model). Thermal model and aging model), service layer (functional modules, such as state estimation, fault diagnosis and residual life prediction) and application layer (advanced applications, such as cascade decision-making, recycling path optimization and carbon footprint calculation). This organic integration of five-layer architecture constitutes the digital foundation to support battery life cycle management.

Digital twin framework has unique advantages in carbon footprint tracking. Traditional life cycle assessment (LCA) methods usually use static and average emission factors, which makes it difficult to reflect the different carbon footprints of specific batteries. The dynamic LCA based on Digital Twins can update the calculation results of carbon footprint in real time according to the actual use conditions of each battery, the carbon emission intensity of the connected power grid and the actual recycling process, which is of great practical significance to meet the trade compliance requirements such as the European Union Carbon Border Adjustment Mechanism (CBAM) and support the enterprise-level carbon neutrality commitment.

6. LIMITATIONS

The review has several limitations that must be acknowledged. First of all, this study did not follow the systematic literature selection scheme. The cited references are selected because of their thematic relevance, rather than by searching the database with clear inclusion and exclusion criteria. Secondly, the quantitative data proposed in this paper, especially the comparative indicators in Table 1, are compiled from heterogeneous literature sources with different experimental conditions, which limits their direct traceability and comparability between studies. Thirdly, all the conclusions drawn here are based on the secondary analysis of published experimental results; There is no original experimental verification in this work. These limitations indicate that the results should be interpreted as a theoretical summary, rather than an empirical statement.

7. CONCLUSIONS AND OUTLOOK

This paper systematically reviews and analyzes the aging mechanisms and green recycling technologies of new energy vehicle power batteries from four dimensions. The main conclusions are as follows: (1) Power battery aging is a complex process driven by multi-physical field coupling; the LLI/LAM decoupling framework provides effective tools for quantitatively identifying aging mechanisms, but real-time diagnosis under dynamic operating conditions remains a technical challenge. (2) Mechanism-data fusion SOH prediction methods achieve a good balance between accuracy and generalization capability and represent an important direction for future BMS algorithm development. (3) At the recycling technology level, hydrometallurgy remains the most mature commercialization solution, while direct repair technology demonstrates tremendous potential in reducing energy consumption and carbon emissions; its industrialization bottlenecks mainly concentrate in raw material sorting and process standardization. (4) Digital twin framework can help manage the whole life cycle of battery, and it is one of its most useful functions to track its carbon footprint in real time.

Looking forward to the future, more research is needed in these areas: First, create a simple and fast algorithm to monitor the health of the battery, which can play a role in the automobile BMS. Secondly, the aging database of various brands and types of large batteries is established to help test and improve the calculation method. Thirdly, the direct maintenance technology is used to carry out large-scale tests to make the whole chain of battery sorting, maintenance and testing. Fourth, find a solution of hybrid blockchain and digital twins to ensure the security of data and share it among all participants during the whole battery life. Advancing this work will help to build a safer, more efficient and better car battery recycling system for our planet.

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