Review of State Estimation and Remaining Useful Life Prediction Methods for Lithium–Ion Batteries

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Abstract: The accurate estimation of the state of charge, the state of health and the prediction of remaining useful life of lithium–ion batteries is an important component of battery management. It is of great significance to prolong battery life and ensure the reliability of the battery system. Many researchers have completed a large amount of work on battery state evaluation and RUL prediction methods and proposed a variety of methods. This paper first introduces the definition of the SOC, the SOH and the existing estimation methods. Then, the definition of RUL is introduced, and the main methods are classified and compared. Finally, the challenges of lithium–ion battery state estimation and RUL prediction are summarized, and the direction for future development is proposed.

Keywords: lithium–ion battery; SOC estimation; SOH estimation; RUL prediction

1. Introduction

Lithium–ion batteries have the advantages of high energy density, low self-discharge and long life, and have been used in many fields [1,2]. Given the background of an expanding lithium–ion battery global market, related technical research such as state-of-charge (SOC) estimation needs to be gradually improved to effectively ensure the promotion and application of lithium–ion batteries in multiple scenarios. Accurate estimation of the SOC plays an important role in preventing battery overcharge, overdischarge and extending the battery cycle life. However, lithium–ion batteries are highly nonlinear, and their SOC is affected by many factors, making it difficult to accurately estimate the SOC. Therefore, the SOC estimation method of lithium–ion batteries is of widespread concern.

In addition, in the continuous charging and discharging process of lithium–ion batteries, battery performance will deteriorate with the decrease in capacity and increase in impedance; this may induce battery faults, such as internal short circuits and thermal runaway, resulting in equipment and system failure and catastrophic accidents [3]. Therefore, it is also critical to accurately estimate the state of health (SOH) and predict the remaining useful life (RUL) to improve the reliability of the battery [4].

At present, some scholars have summarized and solved the estimation and prediction methods of the SOC, the SOH and the RUL. Reference [1] summarized the methods and models for SOH estimation and compared their advantages and disadvantages, but the methods are not well classified or comprehensively covered. Reference [2] provided a coherent review of battery state estimation and RUL prediction techniques and provided an available commercial dataset for analysis; however, the relationship between these methodologies is not clearly explained. Reference [5] reviewed model-based methods and data-driven methods for SOC estimation, but the models of battery are not compared in detail. With the development of the battery industry, there have been a lot of innovations in battery state estimation and life prediction methods. However, previous reviews cannot fully cover innovative methods. Thus, this paper explains and summarizes recent extensive scholarly research.
The structure of this paper is as follows: Section 1 introduces the SOC estimation methodology. Section 2 introduces the SOH estimation methodology. Section 3 summarizes the estimation method of RUL. Section 4 summarizes and suggests possible future research directions based on the existing problems in current research.

2. SOC Estimation Method

2.1. Classification of SOC Estimation Methods

Battery SOC refers to the available state of the remaining charge in the battery. It is the ratio of the remaining charge margin $Q_{\text{remain}}$ of the battery to the nominal (rated) charge capacity $Q_{\text{rated}}$ as shown in Figure 1. Generally, it is expressed as follows:

$$S_{\text{OC}} = \frac{Q_{\text{remain}}}{Q_{\text{rated}}} \times 100\% \quad (1)$$

In this paper, the SOC estimation methods for a lithium–ion battery are summarized into the following four categories [5]:

1. The experiment-based SOC estimation method [6] involves the accurate estimation of the SOC by experimentally measuring the value of cell features;

2. The model-based SOC estimation method [7,8] includes the equivalent circuit model, the electrochemical model and other relevant battery models which are usually employed to estimate the state parameters of the battery’s SOC;

3. The data-driven SOC estimation method [9] fits and combines a large number of data to estimate the battery’s SOC through machine-learning;

4. The fusion of multiple methods [10] aims to balance the accuracy of estimation and reduce the calculation time. These multi-class methods learn from each other.

Figure 2 shows the selection of the estimation methods for the SOC.
2.2. The Experimental SOC Estimation Method

2.2.1. Ampere Integration

Ampere–hour integration (also known as the current integration method or the Coulomb counting method) estimates the SOC of the battery through the accumulated charge and discharge. Compared with other SOC estimation methods, the current integration method is relatively reliable and can dynamically estimate the SOC of the battery, thus, it is widely used in real applications. It is expressed as:

\[
SOC = SOC_0 - \frac{1}{C_N} \int_{t_0}^{t} \eta I dt
\]  

(2)

where \(SOC\) is the state of charge at time \(t\); \(SOC_0\) is the SOC at the beginning time of \(t_0\); \(C_N\) is the nominal capacity of the battery under the current standard conditions; \(\eta\) is the coulomb efficiency; \(I\) is the current, which is negative when charging and positive when discharging. The factors affecting the accuracy of the ampere integral method can be seen from Equation (2):

1. The initial \(SOC_0\) is mainly related to the sampling accuracy and frequency of the current sensor;
2. The Coulomb efficiency \(\eta\) is deeply affected by the working state of battery (e.g., charged state, temperature, current, etc.), and is difficult to measure accurately;
3. The available capacity \(C_N\) is affected by battery aging and operating conditions (charge/discharge ratio, temperature, etc.).
Due to error accumulation, the ampere–hour integration method does not meet requirements for accuracy of SOC estimation. Some scholars have carried out research on improving the accuracy of the ampere–hour integration method. Reference [11] tested the lithium iron phosphate power battery and compared the importance of various parameters for improving the accuracy of SOC estimation. The results showed that the initial SOC correction was the most important for improving the accuracy of the amp–hour integration method. Reference [12] points out that in a simulation process, if the test current is not corrected in time, it will lead to large cumulative measurement errors due to the open-loop working mode of ampere–hour integration. At the same time, reference [12] proposed the ampere–hour integral method with capacity correction for the error caused by the traditional method which treats the available capacity of the battery as a fixed value. Reference [13], focusing on the error caused by the inability to update the Coulomb efficiency online in the traditional amp–hour integration method, proposed an amp–hour integration method to modify the electricity after piecewise integration by adopting various Coulomb efficiencies.

2.2.2. The Open-Circuit Voltage-Based Method

Since the terminal voltage of the battery has a fixed functional relationship with the SOC under the condition of long-term standing, the battery’s SOC can be obtained by measuring the curve corresponding to the open circuit voltage (OCV) and the SOC [14]. When the ampere–hour integration method is used, it often relies on the OCV–SOC curve to calibrate at the beginning/end of charge and discharge. However, there are three problems with the open-circuit voltage method:

1. When measuring open-circuit voltage, the battery needs to be rested for a long time to achieve voltage stability, which is difficult for real-time applications;
2. It is difficult to determine how long the rest time is;
3. As shown in Figure 3, the OCV–SOC curve has a hysteresis effect [15]; the same voltage corresponds to different SOCs when charging and discharging, which will also leads to large errors in SOC calculation.

![Figure 3. OCV–SOC curve of lithium–ion battery.](image)

2.2.3. Other Methods

Electrochemical impedance spectroscopy uses a small amplitude sinusoidal AC signal to perturb the electrochemical power supply system. By measuring the battery impedance at different frequencies, the internal reaction mechanism of the battery that is most relevant to the change in the battery’s SOC is found, so as to estimate the battery’s SOC value [15–17]. Electrochemical impedance spectroscopy can quickly and directly reflect the dynamic characteristics of batteries, and can decouple the factors affecting the SOC. However, it is difficult to measure electrochemical impedance spectroscopy in practical applications.

The load voltage-based method is also commonly used to estimate the SOC. Since the variation law of load voltage on the SOC is similar to that of an open-circuit voltage with SOC, the SOC can be roughly calculated based on voltage variation [18]. This method has
the advantages of simple calculation and high estimation efficiency. However, there is a “platform area” [5] in the relationship curve between the terminal voltage and SOC of the lithium battery. As shown in Figure 4, a certain open-circuit voltage in the platform area corresponds to a wide capacity range, and an inaccurate terminal voltage estimation within this range will lead to a large SOC estimation error. In addition, due to the existence of polarization voltage, the variation in battery terminal voltage is very different when the current changes rapidly, which will also introduce errors in the SOC calculation.

![Figure 4. Curve of terminal voltage SOC for LFP battery. Reprinted with permission from Ref. [19] © 2020 by Zhihu.](image)

2.3. The Model Method

The lithium–ion battery model includes an external characteristic model and an electrochemical model; both can be used for SOC calculation.

2.3.1. SOC Estimation Method Based on the Electrical Model

The method of SOC estimation based on the external characteristic model is relatively simple, involves less computation and is easier to apply in engineering. The electrical model includes the equivalent circuit model and the OCV–SOC models. Commonly used equivalent circuit models include the Rint model, the PNGV model [20], the nth-order Thevenin model [20,21] (nth-order RC model), the GNL model [20], etc. The comparison of different models is listed in Table 1.

<table>
<thead>
<tr>
<th>Classification</th>
<th>Configuration</th>
<th>Description</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rint model</td>
<td>An ideal voltage source is connected to a resistor in series.</td>
<td>The model is simple, and the parameter measurement is easy.</td>
<td>It cannot reflect the dynamic characteristics of the battery and has low accuracy with a small range of applications.</td>
<td></td>
</tr>
<tr>
<td>Electrical model</td>
<td>The n-order Thevenin equivalent circuit model is based on the Rint model; n RC circuits are connected in series to represent the polarization phenomenon of the cell.</td>
<td>The RC loop is used to simulate the dynamic characteristics of batteries, and the higher the n, the higher the precision.</td>
<td>The change in open-circuit voltage and the self-discharge caused by load current accumulated over time are not considered. The bigger the n, the more computation.</td>
<td></td>
</tr>
<tr>
<td>PNGV model</td>
<td>Based on the 1st order Thevenin equivalent circuit model, capacitor Cp is added to describe the change in open-circuit voltage caused by load current over time.</td>
<td>The calculation burden is low; Compared with the 1st order Thevenin equivalent circuit model, the accuracy is higher.</td>
<td>It does not solve the battery self-discharge problem.</td>
<td></td>
</tr>
</tbody>
</table>
The Rint model is the simplest equivalent circuit model, which consists of an ideal voltage source and an internal resistance in series. The model is simple, and the model parameters are easy to measure. However, it cannot reflect the transient process of the battery and has low accuracy and a small application range. Compared with the Rint model, the Thévenin model adds an RC circuit to reflect the polarization process inside the cell. The more RC circuits added, the more accurate the circuit model and the higher the computational complexity and amount. The second-order Thévenin model is also known as the dual polarization (DP) model, in which one RC circuit represents the electrochemical polarization of the battery, and the other represents the concentration polarization of the battery. However, the Thévenin model does not consider the change in open-circuit voltage and self-discharge caused by load current over time. Compared with the first-order Thévenin model, the PNGV model adds a capacitor in the loop to describe the change in the open-circuit voltage with the current integral. The GNL model integrates the advantages of...
the above three models. It not only reflects the ohmic polarization, the electrochemical polarization and the concentration polarization processes of the battery, but also solves the problem of the open-circuit voltage affected by the integral load current and the self-discharge of the battery. It has a wide range of applications, but the complexity of the model and the amount of calculation increase at the same time.

The estimation method based on the OCV–SOC curve can also estimate the lithium-ion battery’s SOC. The main fitting models [22] include the Shepherd model (Equation (3)), the Unnewehr universal model (Equation (4)), the Nernst model (Equation (5)) and the compound model (Equation (6)):

\[ U_k = E_0 - R_i - K_0 \frac{1}{S_{OC_k}} \] (3)

\[ U_k = E_0 - R_i - K_1 S_{OC_k} \] (4)

\[ U_k = E_0 - R_i + K_2 \ln(S_{OC_k}) \] (5)

\[ U_k = E_0 - R_i - K_0 \frac{1}{S_{OC_k}} - K_1 S_{OC_k} + K_2 \ln(S_{OC_k}) + K_3 \ln(1 - S_{OC_k}) \] (6)

where \( U_k \) is the terminal voltage of the battery at time step \( k \); \( i_k \) is the current at time step \( k \); \( E_0 \) is the open circuit voltage after the battery is fully charged; \( R \) is the internal resistance of the battery; \( S_{OC_k} \) is the SOC of \( k \)th time step; \( K_0, K_1, K_2 \) and \( K_3 \) are the parameters to be fitted. A detailed description and comparison of the above models are shown in Table 1.

When estimating SOC based on the electrical model, the battery system is usually discretized, the SOC is taken as the system state and the SOC is estimated using a filter based on the ampere integration method.

Taking the 1st order Thevenin equivalent circuit as an example, as shown in Figure 5, the discretized equation of state and the output equation are shown in Equations (7) and (8), respectively:

\[
\begin{bmatrix}
U_{1,k} \\
S_{OC_k}
\end{bmatrix}
= 
\begin{bmatrix}
\exp(-\frac{I_k}{R C_1}) & 0 \\
0 & 1
\end{bmatrix}
\begin{bmatrix}
U_{1,k-1} \\
S_{OC,k-1}
\end{bmatrix} + 
\begin{bmatrix}
w_{1,k-1} \\
w_{2,k-1}
\end{bmatrix}
\]

\[ [U_k] = U_{oc}(S_{OC_k}) - U_{1,k} - R_0 I_k + v_k \] (8)

where \( U_{1,k} \) is the voltage of the battery polarization capacitor; subscript \( k \) is the current time; \( T_s \) is sampling time; and \( w_k \) and \( v_k \) are system noise and observation noise, respectively. They are Gaussian white noise with mean is zero, covariance \( Q \) and covariance \( R \).

\[ U_{oc}(S_{OC_k}) \] is the function of open circuit voltage \( U_{oc} \) with respect to SOC, which can be obtained by fitting the curve of OCV–SOC. \( R_1, C_1 \) are the internal resistance and capacitance of polarization, respectively. \( R_0 \) is the ohmic internal resistance of the battery; \( Q_1 \) is the actual total capacity under the current \( I \). The battery’s SOC can be estimated by combining the filter and its derived algorithm.

The commonly used filters include the Kalman filter (KF), the particle filter (PF) and the H infinite filter (HIF).
The KF can use the output data to continuously revise the system state variables and provide the optimal estimation of the state quantity at the next moment [23]. The KF is widely used in the research of battery SOC because of its good resistance to noise and low dependence on an initial value.

Since lithium-ion batteries are nonlinear systems, the extended Kalman filter (EKF) is used to estimate the SOC [24], which solves problems caused by system nonlinearity by performing a Taylor expansion at the operating point.

According to Equations (7) and (8), the error of the EKF comes from the following three points:

1. Only the first-order term is taken when the Taylor expansion is carried out on the nonlinear system at the operating point, and the higher-order term is ignored;
2. Noise w_k and v_k, and their covariances Q and R, are constants and do not change dynamically;
3. Inaccurate battery model parameters of R_0, R_1 and C_1 will lead to large errors.

For problem (1), the unscented Kalman filter (UKF) is employed [25]. The UKF eliminates the larger error caused by the partial derivative of the first order using the Taylor expansion of the nonlinear function in the EKF. However, this method needs accurate statistical characteristics for the system process noise and the observation noise. In addition, because the calculation process is based on a matrix, when there are drastic fluctuations of voltage and current in the system, the matrix cannot guarantee strict positive quality, resulting in divergence of state estimation and unstable estimation [26]; the amount of calculation is also large. To solve the problem of the UKF, some scholars proposed the square root unscented Kalman filter (SR–UKF) and ensured the semi-positive nature of the covariance matrix by Cholesky decomposition factor updating and matrix QR decomposition, which increases the stability of calculation [27]. However, problem (2) still remains.

The adaptive extended Kalman filter (AEKF) method was proposed by reference [28] to solve problem (2). Based on the EKF, the AEKF focused on the problem of filtering divergence caused by the unknowability of the covariance matrices Q and R. A forgetting factor is added to the EKF, and the covariance of error is corrected by the observed value. The convergence rate is improved by dynamic changes in Q and R. However, the AEKF does not consider the change in model and equation parameters, and the fixed forgetting factor will also introduce errors.

The idea of the PF is based on the Monte Carlo method, which uses sets of particles to represent probabilities, and can be applied to any form of state–space model. Compared with the KF, the PF is a nonlinear and non-Gaussian filtering method, which does not make a linear assumption or a Gaussian hypothesis of posterior probability. In reference [29], the PF was used to estimate the SOC of Fe–phosphate lithium–ion batteries, and the results showed that it could well describe the external characteristics of nonlinear systems and significantly improve the prediction accuracy of the SOC. However, particle degradation was prone to occur. To solve problems such as particle scarcity and noise disturbance, reference [30] proposed an improved estimation algorithm, the unscented particle filter (UPF), to achieve accurate SOC estimation. However, the UPF requires high computing ability for hardware devices.

The HIF method is an improved algorithm based on the EKF. In the HIF, Q and R are parameters designed according to the prior knowledge of w_k and v_k. Compared with the EKF and the PF, model inaccuracy and noise uncertainty can be better tolerated [31,32]. In reference [33], the HIF algorithm was used to obtain the SOC estimation results with high accuracy and robustness. Reference [34] uses a dual HIF algorithm to estimate the SOC, which can stabilize the estimation error within 2%. Most of the HIF algorithms can achieve high accuracy; however, due to their robustness, there is still a problem of insensitivity to system state and model uncertainty to noise. In reference [35], a nonlinear observer based on HIF was proposed to estimate the SOC for lithium–ion batteries. Experimental results show that this algorithm can suppress model errors under different working conditions, but the algorithm is slow to respond to changes in model parameters.
Similar to filters, observer-based methods can also observe model states in real time by calculating systematic errors [36]. Among them, the sliding mode observer (SMO) is a closed-loop state observer, and the output variable provides correction for the observer system. Due to its closed-loop characteristics, the SMO has strong robustness, low sensitivity to battery model structure and parameter disturbance [37], no prior information to predict noise [38] and its estimation accuracy is close to that of the PF. Reference [39] proposes an SOC estimation method for lithium–ion batteries based on an improved SMO, which inherits the good robustness of the SMO.

2.3.2. SOC Estimation Method Based on the Electrochemical Model

Although the electrical model is simple, it cannot fully reflect the state change caused by the change in the internal mechanism of the battery. The electrochemical model of the lithium–ion battery describes the chemical reaction inside the battery and has a more definite physical meaning. The battery electrochemical model mainly includes a pseudo two-dimensional (P2D) model [40], a single particle (SP) model and a simplified P2D model. The comparison of the three models is also shown in Table 1.

The P2D model regards the lithium battery as a structure composed of electrodes (positive and negative), a diaphragm and an electrolyte full of numerous spherical solid particles. A series of partial differential equations (PDE) is used to describe the dynamic mechanism inside the battery. It can carry out accurate battery state estimation and has universality and scalability. It is suitable for batteries of different material systems and can be developed and extended to more complex multi-field coupling models. However, for complex models, in addition to excessive parameters, it is also difficult to find analytical solutions for PDE [8].

The SP model is the simplest electrochemical model of the lithium battery, which is simplified from the P2D model. In the SP model, two spherical particles are regarded as the positive and negative electrodes of the battery, respectively. It is assumed that the embedding and ejection process of lithium ions occurs on the spherical particles, and the concentration of the electrolyte and its internal potential are considered constant. The single particle model is simple in structure and small in calculation. However, the model assumption is not valid under the conditions of high rates of charge and discharge, so it will introduce large errors.

Since the equations of the P2D model are too complex and the accuracy of the SP model is poor, many scholars have proposed different simplified methods for the P2D model in different application scenarios to ensure calculation accuracy and reduce the amount of calculation. The existing simplification methods mainly include geometric structure simplification, solid–liquid diffusion process simplification and transformation simplification by mathematical algorithm. In reference [41], mathematical equations were established to describe the internal physical and chemical behaviors of power batteries during charging and discharging based on the porous electrode theory and the concentrated solution theory; the finite analysis method was used to grid and discretize the PDE, reducing its order to an ordinary differential equation. However, the simplified model cannot solve the inherent problem of excessive P2D parameters and will reduce the model accuracy to varying degrees. It is still worth thinking about how to balance the amount of calculation and the model’s accuracy [42–45].

When calculating the SOC based on the electrochemical model, different from Equation (1), the SOC of the electrochemical mechanism model (such as the P2D model and the SP model) is divided into surface SOC and average SOC [46], which can be expressed as

\[
\text{SOC}_{\text{surf}} = \frac{\theta_{s,i} - \theta_{s,0\%}}{\theta_{s,100\%} - \theta_{s,0\%}}
\]

\[
\text{SOC}_{\text{bulk}} = \frac{\theta_{s,i} - \theta_{s,0\%}}{\theta_{s,100\%} - \theta_{s,0\%}}
\]
where \( S_{\text{OCsurf}} \) and \( S_{\text{OCbulk}} \) represent surface SOC and average SOC, respectively. The parameters \( \theta_{s,i} \) and \( \theta_{s,i}^{\text{avg}} \) represent electrode utilization and the average utilization rate of the electrode, respectively, while \( \theta_{s,i}^{\text{avg}} \) is the ratio of solid lithium ion concentration to the largest solid lithium ion concentration ratio. The electrode utilization ratio of \( \theta_{s,0\%} \) and \( \theta_{s,100\%} \) is for 0% SOC and 100% SOC, respectively.

Similar to the calculation of the SOC based on an external characteristic model, the SOC of an electrochemical model can be estimated by a filter or observer.

2.4. Data-Driven Methods

The relationship between battery parameters and the SOC is complex and non-linear, making it difficult to establish a model with traditional mathematical methods [47]. Data-driven methods do not rely on a specific model of the battery, and can directly rely on the mapping relationship between the input and output of the system to estimate the SOC. At present, widely used data-driven methods include neural networks, regression analysis, etc.

2.4.1. Neural Networks and their Improvement Method

The SOC estimation method based on neural networks takes the battery operating data, such as voltage, current and other parameters as input and SOC as output and trains the system through sample data to find the mapping relationship between various parameters [48].

The basic structure of a neural network (NN) consists of a three-layer formation as shown in Figure 6. The input layer takes the vector of instantaneous current, voltage and temperature values. The output layer is the instantaneous SOC value. By training the NN, it is able to form a non-linear map that accurately models the input–output relationship without any prior knowledge of the internal structure of the battery [49]. The relationship between the input layer and the output layer is developed using a suitable number of hidden layers, hidden neurons and an activation function. The SOC in the output layer can be expressed by

\[
SOC_i = f_i \left\{ \sum_k w_{i,k} a_j + \theta_{j,k} \right\}
\]

(11)

![Figure 6. The general architecture of the 3-layer neural network for SOC estimation. Reprinted with permission from Ref. 49 © 2018 by Elsevier.](image)

A back propagation (BP) neural network is a typical neural network algorithm. Its network structure is simple, and it has a strong nonlinear mapping ability. Reference [50] summarized the influencing factors of SOC estimation and proposed a method of SOC
estimation based on a BP neural network. However, BP neural networks tend to easily converge to local optimum. In addition, the network structure is different, making it easy for non-convergence to appear. Currently, there exists a number of common NN architectures. Recurrent neural network (RNN) is a neural network that takes sequence data as input and makes recursion in the evolution direction of the sequence; all loop nodes are connected by a chain, which is suitable to estimate the SOC and other temporal data [51]. However, during its training, gradient vanishing and gradient explosion will appear. To solve these two problems, some scholars propose a long short-term memory (LSTM) neural network [52–54], a gated recurrent unit (GRU) neural network [55,56] and a Bi LSTM neural network [57]. LSTMs solve the problem of gradient vanishing and gradient explosion during long sequence training. Compared to an ordinary RNN, an LSTM can perform better in longer sequences. A GRU is an improvement on an LSTM, with the same accuracy as LSTM, but with fewer parameters and faster computation. In reference [58], an RNN with the GRU is established to evaluate the SOC for a LIB. The ensemble optimization method based on a Nadam and AdaMAx optimizer is used to improve the training operation and determine the optimal parameters.

However, all neural networks and their derivative methods have three disadvantages: they need to collect a large amount of data, which is not applicable for online estimation; training results are too dependent on sample quality; and the overfitting problem appears easily since the battery’s model is not involved.

2.4.2. The Regression Analysis Method and Its Improvement

The regression analysis method involves processing a large number of statistical data to determine the correlation between dependent variables and some independent variables. It establishes a regression equation with good correlation and can be extrapolated to predict the future change in dependent variables.

A support vector machine (SVM) is an algorithm that uses classification and regression analysis techniques to process data [59]. This method is effective in high-dimensional pattern recognition and nonlinear regression problems. An SVM attempts to construct hyperplanes in high-dimensional space in order to separate data of one class from another as shown in Figure 7 [60]. An optimal separation boundary is achieved when the distance from the hyperplane to the nearest data point of any class is maximized. Reference [61] employed SVM to predict the SOC of a LIB as a function of cell current, voltage and temperature. However, when the sample size is expanded to a certain point, the complexity brought by the SVM optimization will be significantly increased, and the accuracy of the model will decrease [62,63].

![Figure 7. Example of hyperplane to separate distinct classes for the SVM. Reprinted with permission from Ref. [60] (© 2005 by Springer).](image)

To solve the above problems, reference [62] proposed a least squares SVM, which converts the learning problem of the standard SVM into a problem of solving linear equations with fewer variables and less computation, with faster solving speed and better robustness.
2.5. The Joint Method

Due to the advantages and disadvantages of each method, some scholars use the integration of multiple methods to estimate the SOC, with complementary advantages. The fusion types include the fusion of a model-based method and a data-driven method, the fusion of different data-driven methods, etc. Reference [64] proposed a convolutional neural network (CNN) and an LSTM integration model, which established a mapping relationship between the SOC at the current moment and the discharge data of lithium batteries at several historical moments, improving the accuracy of SOC estimation. Reference [65] analyzed the nonlinear relationship between the OCV and the SOC based on the Thevenin equivalent circuit model and combined the models, employing improved UKF with neural networks to estimate the SOC; this was more accurate than the ordinary UKF.

After a comprehensive analysis of the above methods for estimating the lithium–ion battery’s SOC, the advantages and disadvantages of common methods for estimating the lithium–ion battery’s SOC are compared, as shown in Table 2.

<table>
<thead>
<tr>
<th>Classification</th>
<th>Method</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experimental method</td>
<td>Ampere integration</td>
<td>Non-relative internal mechanism of the battery; simple</td>
<td>Easy to generate cumulative error, which requires high initial value and sensor precision</td>
</tr>
<tr>
<td>OCV-SOC method</td>
<td>Electrical model</td>
<td>Simple and practical</td>
<td>Poor accuracy</td>
</tr>
<tr>
<td>Basic model</td>
<td>Electro-chemical model</td>
<td>It reflects the internal characteristics of the battery</td>
<td>Complex and computationally intensive [5]</td>
</tr>
<tr>
<td>State estimation method</td>
<td>KF method</td>
<td>The convergence speed is fast, and the noise suppression ability is strong, Low sensitivity to initial value</td>
<td>The system noise is uncertain, which requires high accuracy of the model</td>
</tr>
<tr>
<td>PF method</td>
<td>Strong robustness and low requirement for model accuracy</td>
<td>Prone to particle degradation</td>
<td>Computationally intensive</td>
</tr>
<tr>
<td>Data-driven method</td>
<td>Neural network</td>
<td>No reliance on high-precision battery models</td>
<td>Easy to disappear gradient and fall into local optimization</td>
</tr>
<tr>
<td>Regression analysis</td>
<td>It can achieve good results in high-dimensional pattern recognition, nonlinear regression and other problems</td>
<td>Only applicable to small data samples</td>
<td></td>
</tr>
<tr>
<td>Joint method</td>
<td>Model-based method and data-driven method</td>
<td>The accuracy and reliability of the estimation results are high</td>
<td>High complexity and computationally intensive [2]</td>
</tr>
</tbody>
</table>

3. RUL Prediction Method

3.1. Classification of RUL Prediction Methods

Due to the electrochemical side reaction in the anode and cathode electrolyte of the battery, as well as the influence of mechanical stress, manufacturing process defects, environmental temperature and other external factors, the degradation of batteries has become a complex problem [66]. Determining the RUL of a battery under different operating conditions is of great significance to accurately estimate the battery replacement cycle or maintain the battery [67]. It is generally believed that when the real-time capacity of lithium–ion battery drops to 80% of the rated capacity, the critical value of battery failure is reached [68].

The RUL refers to the number of charge/discharge cycles before the battery health state has deteriorated to a point where it reaches EOL [69]. As Figure 1 shows, the RUL is the remaining cycle numbers of the battery from the current SOH to EOL. Thus, the SOH and the RUL can be estimated with the same methodology. The SOH estimation methods are not further reviewed in this paper. Similar to the SOC estimation, the RUL estimation and prediction methods for the lithium–ion battery comprise a model-based method, a...
data-driven method and a joint method since the RUL can also be regarded as a battery state. Different from the SOC, the RUL is not an instantaneous characteristic so it can be predicted through an empirical model, a semi-empirical model and a time series prediction data-driven method.

3.2. The Model-Based Method

As Figure 8 shows, according to different modeling mechanisms, that the model-based method can be categorized into an empirical model, a semi-empirical model, an electrical model and an electrochemical model.

Figure 8. Classification of RUL prediction methods for a lithium–ion battery.

3.2.1. Empirical Model

In empirical models, battery capacity attenuation is represented as a mathematical function of number of cycles. Different mathematical functions are used to describe the battery capacity deterioration trend. Most empirical models for RUL estimation are developed based on a single exponential model [70], a double exponential model [71], a linear model [72], a polynomial model [73], a Verhulst model [74], etc.

\[
\text{single exponential model: } C_{\text{max}} = a_1 \exp(a_2 n) + a_3
\]  

\[
\text{double exponential model: } C_{\text{max}} = b_1 \exp(b_2 n) + b_3 \exp(b_4 n)
\]  

\[
\text{double exponential model: } C_{\text{max}} = b_1 \exp(b_2 n) + b_3 \exp(b_4 n)
\]  

\[
\text{linear model: } C_{\text{max}} = c_1 n + c_2
\]
polynomial model: \[ C_{\text{max}} = d_1 n^2 + d_2 n + d_3 \] (16)

Verhulst model: \[ C_{\text{max}} = \frac{e_1 / e_2}{1 + [e_1 / (e_2 C_0) - 1] \exp(e_1 n)} \] (17)

where \( n \) is the equivalent cycle number; \( C_{\text{max}} \) is the remaining capacity at \( n \)th cycle; and \( a, b, c, d, e \) are constant coefficients to be fitted.

The empirical model only needs one function to evaluate the RUL, which is easy to operate and small in calculation. However, due to its lack of real physical meaning, the estimation accuracy is low.

### 3.2.2. The Semi-Empirical Model

The semi-empirical model considers the influence of various physical factors on battery aging by adopting the stress factors related to battery aging and modeling the change in stress factors. Stress factors related to battery aging are usually modeled based on the inverse power law [75], the Arrhenius model [76] and the double exponential model. The stress factors can be classified according to different criteria:

1. Battery parameters including the SOC [77], solid electrolyte interphase (SEI) film formation and capacity plummeting [78], etc.;
2. Operating conditions including charge or discharge rate [74], charge or discharge cut-off voltage [79], depth of discharge (DOD) [80,81], Ampere hour (Ah) throughput [82], etc.;
3. Operating environment including temperature [81], etc.

Different stress factors can be coupled to obtain the comprehensive capacity decline rate to redevelop a basic empirical model. Reference [83] applied a semi-empirical model related to normal temperature, abnormal temperature and discharge rate to evaluate the RUL. The result showed high accuracy with an estimation error of 2.22%. Besides discharge rate and temperature, reference [84] coupled the stress factor of cut-off voltage, the discharge rate and temperature to simulate the battery aging process. Reference [85] established a capacity decline model based on the SEI films formation mechanism. However, the semi-empirical model is no longer applicable when the operating conditions of the battery change significantly since the variation in stress factors cannot be modeled [85].

### 3.2.3. The Electrical Model

Similar to the SOC estimation, the RUL can also be regarded as a battery state. Thus, model-based methods for SOC estimation can also be employed to predict the RUL. Reference [86] proposed an improved UPF to predict the RUL with an error of less than 5%. Reference [87] proposed a new PF framework based on a conditional variational self-coder for RUL prediction of lithium batteries. In the process of particle resampling, a reweighting strategy is introduced to prevent the loss of particle diversity. Compared with the traditional method, this method improves the prediction accuracy.

### 3.2.4. The Electrochemical Model

The electrochemical model describes the influence of internal chemical and physical reactions on battery aging, such as electrical conductivity loss, activated substances loss and lithium–ion loss. However, the complexity of the electrochemical model and poor dynamic accuracy limit its application in RUL prediction with a large number of aging related parameters to be identified as compared to the electrical model [88]. Reference [89] employed the mixtures of the inverse Burr distribution with inverse Gaussian and inverse Weibull distributions to analyze a battery dataset and predict the RUL through the Monte Carlo method.

### 3.3. The Data-Driven Method

The data-driven method selects some historical data as training samples, and then uses the training algorithm to predict the RUL after training. This method avoids the complex
mathematical model establishment and expertise required for traditional methods. It can be roughly classified as a time series prediction method and feature-based method.

3.3.1. The Time Series Prediction Method

The time series prediction method predicts future capacity directly based on the change trend of historical capacity time series data.

The regression model, NN and entropy-based methods are usually employed to predict using time series. Reference [90] enhanced a PF-based empirical method by introducing a NN to model battery degradation trends under various operation conditions to improve their generalization and nonlinear representing ability, and recursively update the NN model’s parameters with a particle filter to reduce the degeneracy and impoverishment of the PF. Reference [91] proposed an RUL prediction method based on an autoregressive integrated moving average (ARIMA) model. However, ARIMA model requires the stability of the time series data and has high requirements for battery operating conditions. Reference [92] constructed a CNN model for RUL prediction with an accuracy of over 90%. However, gradient disappearance is a typical problem for all NN methods. The following problems of the time series prediction method still require solution:

1. Low accuracy for local capacity variation;
2. Not applicable to predict the RUL in the battery’s entire life cycle due to different degradation characteristics in each aging period;
3. High dependence on the training set and data quality;
4. Problem of overfitting.

3.3.2. The Feature-Based Method

The feature-based method improves the robustness of the basic time series prediction method by considering health factors (HFs) which indicate battery aging. In comparison to direct time series prediction, the feature-based method has a better performance in predicting the RUL curve’s local fluctuations (e.g., the capacity regeneration effect).

The procedure for the feature-based data-driven method is summarized as data acquisition, data preprocessing, feature extraction, feature election and data-driven prediction, as shown in Figure 9.

![Figure 9. Procedure for feature-based data-driven method.](image-url)

The battery HFs extracted are mainly derived from the voltage, current and temperature curves. The HFs that have appeared in other researches are summarized in Table 3 [93–98]. Then, feature selection is conducted if necessary. Since there are various HFs related to battery aging and the HFs are coupled together, in order to reduce the calculation burden, some researchers analyze the correlation of the HFs with the RUL and select the most relevant HFs through ranking. The Pearson coefficient, the Spearman coefficient and gray relation analysis are usually used to analyze the correlation between health factors and capacity attenuation. In reference [95], HFs that are highly correlated with the capacity degradation curve are chosen by means of gray relation analysis and used to predict the RUL with GPR. Dimension reduction means are also employed to reduce the
size of HFs series to figure out the most relevant part. Typical dimensionality reduction methods include Principal Component Analysis (PCA) [96], Independent Component Analysis (ICA) [96] and autoencoder [96]. Moreover, the performance of raw HFs may not be fully satisfactory due to the complexity of the underlying degradation process. To reduce the possible deviation of the HF from the degradation process, the Box–Cox transformation is sometimes used to enhance the linear relationship between the HF and the system’s health status [99,100].

Table 3. Commonly used health factors of lithium–ion battery.

<table>
<thead>
<tr>
<th>Classification</th>
<th>Health Factors</th>
</tr>
</thead>
<tbody>
<tr>
<td>Voltage curve relevant</td>
<td>Constant voltage charging time [93], voltage increment in the same time interval [94], time consumed for certain voltage increments [95], dQ/dV [97], dV/dQ [97], maximum slope of voltage curve [95]</td>
</tr>
<tr>
<td>Current curve relevant</td>
<td>Constant current charging time [93], current increment in the same time interval [94], time consumed for certain current increments [96], area under current curve [95]</td>
</tr>
<tr>
<td>Temperature</td>
<td>Temperature increment in the same time interval [94], time consumed for certain temperature increments [95]</td>
</tr>
<tr>
<td>Other</td>
<td>Wavelet packet energy entropy [98]</td>
</tr>
</tbody>
</table>

Typically, the NN, the regression method and their improvement are employed to predict the RUL with an indicator of battery HFs. Reference [101] trained the RNN model with constraint derived from prior physical knowledge to forecast the RUL. Reference [58] employed a hybrid method with a PF and GPR to predict the aging trend sequence and residual sequences. The proposed approach showed wide generality and reduced errors. Reference [69] optimized the deep learning neural network (DLNN) with evolutionary algorithms to predict the RUL.

3.4. The Joint Method

The joint method is a combination of the model-based method and the data-driven method and the combination of various data-driven methods that fuses the merits of both methods. To improve prediction accuracy, reference [102] proposed an SVR-based model with the approach of the normal particle swarm optimization (PSO). Reference [103] proposed a joint method extreme learning machine (ELM), grey wolf optimization (GWO) and differential evolution (DE). Reference [104] proposed a neural network-based method combining LSTM with PSO and an attention mechanism for RUL prediction [105]. However, all joint methods share the demerits of high computational complexity and uncertainty.

The RUL prediction methods are compared in Table 4.

Table 4. Comparison of lithium–ion battery RUL prediction methods.

<table>
<thead>
<tr>
<th>Classification</th>
<th>Method</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Model-based method</td>
<td>Empirical model</td>
<td>Low calculation burden [3]</td>
<td>Poor accuracy; not be able to consider influence of working conditions and the environment [2]</td>
</tr>
<tr>
<td></td>
<td>Semi-empirical model</td>
<td>Able to consider influence of working conditions and environment</td>
<td>Low accuracy; low robustness under dramatic working condition changes [1]</td>
</tr>
<tr>
<td></td>
<td>Electrochemical model</td>
<td>Able to reveal the aging mechanism</td>
<td>Large calculation amounts due to too many parameters [3]</td>
</tr>
</tbody>
</table>
### Table 4. Cont.

<table>
<thead>
<tr>
<th>Classification</th>
<th>Method</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Data-driven method</td>
<td>Time series prediction method</td>
<td>Does not require complex mathematical models</td>
<td>Large calculation amounts; overfitting; unable to reveal local RUL variation</td>
</tr>
<tr>
<td></td>
<td>Data-driven method and data-driven method</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### 4. Conclusions

This paper summarized the techniques, models and algorithms used in lithium–ion battery state estimation and RUL prediction in recent years and provides a comparison of various methods. The actual operating conditions and environment of lithium–ion batteries vary greatly. Batteries are highly complex, dynamic and nonlinear electrochemical systems, which impose some challenges in practical application for the existing lithium–ion battery SOC estimation methods and RUL prediction methods.

The engineering application capability of existing SOC estimation methods is poor. It is difficult for an experimental method to estimate the SOC during battery operation, and the application of the model-based method is limited because it relies heavily on the accuracy of the battery model.

Some RUL prediction methods have low capability for generalization. In terms of the empirical model and semi-empirical models, model accuracy is greatly affected by battery parameters and working conditions, so they are difficult to generalize in real practice. In terms of the data-driven method, the training data set is all the experimental data obtained under a single working condition, which makes it difficult to use for covering complex scenarios. The parameters of the data-driven method are usually fixed on most occasions, hence it is easy to overfit when applied to other battery models and conditions.

1. The balance between calculation time and accuracy is difficult to reach. Due to the limitations in the number of computations required, most algorithms are not suitable for online application. In addition, since batteries are usually integrated as modules, calculations multiply in the real world which makes real-time application a challenge;
2. There are only relevant studies on single batteries at present; however, factors such as the inconsistency of batteries in battery strings in large-scale lithium battery systems such as in an energy storage power station, will cause the original method to no longer be applicable. At present, there is little research on the state estimation and RUL prediction methods of battery packs.

These problems make most of the lithium–ion battery state estimation and the RUL prediction methods difficult to promote on a large scale, but these challenges also show the future direction for further research on state estimation and RUL prediction of lithium–ion battery:

1. Develop the pack-level, cluster-level, system-level battery equivalent model to reduce the amount of calculation required for state evaluation and RUL prediction;
2. Enhance state estimation and RUL prediction methods with joint algorithms to compensate for weaknesses and improve the algorithm’s capacity for generalization;
3. Establish a multi-physical field coupling model for lithium–ion batteries to improve the accuracy of battery status assessment and RUL prediction.

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