Journal of Cleaner Production 261 (2020) 120813

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Review

A review of the state of health for lithium-ion batteries: Research status and suggestions

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A R T I C L E I N F O

Article history:
Received 20 November 2019
Received in revised form
25 February 2020
Accepted 27 February 2020
Available online 13 March 2020

Handling Editor: Prof. Jiri Jaromir Klemeš

Keywords:
Battery electric vehicles
Lithium-ion battery
State of health
Battery aging
Prediction method

A B S T R A C T

Lithium-ion batteries (LIBs) have become the mainstream power source for battery electric vehicles (BEVs) with relatively superior performance. However, LIBs experience battery aging and performance degradation due to the external environment and internal factors, which should be reflected in the evaluation of the state of health (SOH). Accurately predicting SOH can improve the overall life of the battery and support safe driving in BEVs. At present, while there are many prediction methods for SOH, most are implemented in simulated environments but are challenging to execute in actual industrial production. This review provides a discussion on the aging reasons for LIBs, introduces the SOH prediction method based on the classification framework, and analyzes the key benefits and drawbacks of each method. Finally, the corresponding suggestions and solutions are given in combination with the actual industrial production.

Contents

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https://doi.org/10.1016/j.jclepro.2020.120813
0959-6526/

1. Introduction

Alongside continued advancements in science and technology, energy and environmental crises have increasingly become prevalent and catastrophic. Developments in new energy vehicles (NEVs) have become the preferred alternative for sustainable development. LIBs, the primary power source of NEVs, have been widely used in communication, aviation, automobile, and other industrial fields. Its main advantages are high output voltage, longevity in the life cycle, high energy density, low self-discharge rate, and wide operating temperature range. Some popular examples of LIBs include Chevrolet Volt, Nissan Leaf and BYD E6, NASA's Spirit (Code MER-A), and Opportunity (Code MER-B) Mars Detector, Phoenix Mars Lander, ESA's Mars Express.

However, aging and improper operation of LIBs can significantly affect performance. When the battery performance degrades to a certain extent, battery leakage, insulation damage, and partial short circuit problems can cause catastrophic accidents. For instance, a Tesla Model S suddenly caught fire in an underground garage in Shanghai on April 21, 2019. A Samsung Galaxy S10 mobile phone was damaged by self-ignition during charging in 2019. After investigation, these accidents have been linked with the aging of LIBs. Therefore, research on the state of health (SOH) of LIBs is essential to avoid disastrous accidents caused by aging and performance degradation of LIBs.

At present, some people describe the battery aging and performance from the remaining useful life (RUL), so SOH and RUL are closely linked and can be both used to characterize battery performance. Numerous studies have been published for SOH and RUL and are summarized in Table 1.

Several major limitations can be found in the current literature in SOH/RUL prediction, as highlighted in Table 1. This article proposes recommendations to solve some of the problems and improve the current literature. So, the main objective and the problems tackled in this article are shown in Fig. 1.

This review would be useful for automobile engineers and experts to better understand the current SOH prediction methods and trends. In addition, this article would be able to contribute to future improvements in NEVs. This review is divided into 8 sections. Sections 2 describes how the review is implemented. Section 3 introduces the classification and development trends in NEVs, the LIBs of battery electric vehicles (BEVs), and the main factors affecting LIBs performance and life deterioration. Section 4 discusses the definition and relationship between the SOH and RUL. Section 5 provides a detailed and comprehensive introduction to the current SOH prediction methods and frameworks based on data-driven models, hybrid methods, and other techniques. Section 6 evaluates the proposed SOH prediction methods that combine actual industry data. Section 7 details the current situation and provides recommendations that utilize actual BEVs. The final section provides the summary of this review article.

2. Review screening methods

This review is conducted based on content analysis. The Google Scholar, Baidu Scholar, Web of Science, EI Village were used to find relevant articles for this review. Our review process was carried out using sub-modules, and the summaries of previous modules are provided in the end. In searching for relevant articles, we used joint keywords, such as electric vehicle and lithium-ion battery, lithium-ion battery and aging, lithium-ion battery and health status, lithium-ion battery and SOH, and lithium-ion battery and RUL. We found 875 articles from our initial search. Then, we trimmed the number to 300 articles by analyzing the title, abstract, keywords, content, and the main topics of interest of the journal. In the end, we selected 134 articles in our final list based on the impact factor, citation count, and review process. The schematic diagram is shown in Fig. 2.

Table 1
Evaluation of a few review papers.

Literature	Content	Drawbacks
Literature (Ungurean et al., 2017)	Introduced the prediction methods of SOH and RUL for LIBs and evaluated from six aspects.	Classification confusion of SOH prediction methods.
Literature (Lipu et al., 2018)	Introduced comprehensively the prediction method of SOH and RUL; the evaluation and several suggestions were also offered.	Some of the SOH and RUL methods mentioned in this paper can be summarized as the same type of prediction. It is not stated separately the temperature in the challenge of SOH prediction.
Literature (Barre et al., 2013a)	Introduced the research and progress of the aging mechanism of LIBs in recent years and summarized the SOH prediction method based on the equivalent circuit model (ECM).	Some methods based on the ECM can be combined.
Literature (Farmann et al., 2015a)	Evaluated the prediction methods of SOH/RUL from the side by way of assessing the battery of the available capacity.	Some methods are still not fully described.
Literature (Zhang and Lee, 2011)	Introduced the methods used in the state of charge (SOC) estimation, voltage estimation, capacity estimation, and RUL prediction.	Data-driven approaches are still not covered in depth.

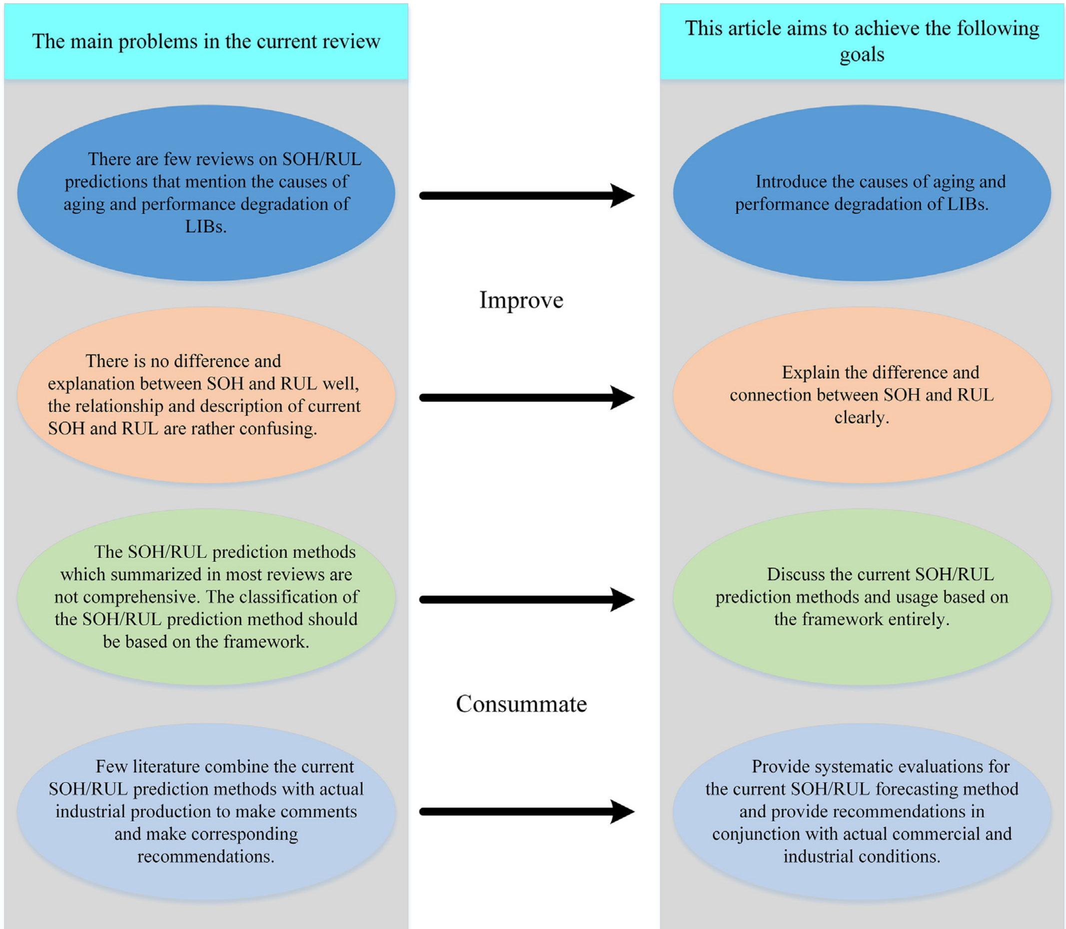


Fig. 1. The current review issues and the goals of this review.

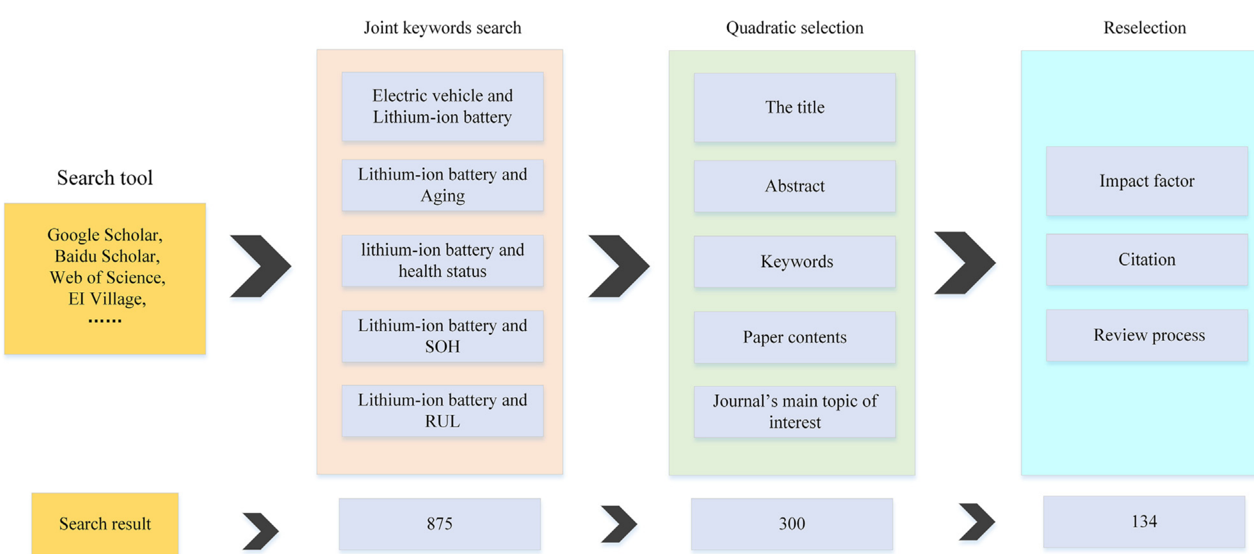


Fig. 2. The review screening methods schematic diagram.

3. Power battery of BEVs and battery aging

3.1. The BEVs in NEVs

NEVs have received extensive attention in various countries and is considered as an emerging strategic industry (Kendall, 2018). This includes automobiles that integrate advanced technology in power control and driving, and form a new type of vehicle having advanced principle technology. NEVs cover a wide range and can be divided into three main categories: hybrid electric vehicles (HEVs), fuel cell electric vehicles (FCEVs), and BEVs. Among them, the HEVs can be further subdivided into plug-in hybrid electric vehicles (PHEVs), range-extended electric vehicles (REEVs), and ordinary HEV. A brief comparative analysis of the different types of NEVs is shown in Fig. 3.

At present, NEVs can solve the pressing problems of environmental pollution and resource shortage by using clean and pollution-free energy. And while the most significant advantage of HEVs is its efficient fuel economy, HEVs can be quite expensive. The FCEVs have the advantages of the BEVs and without any associated pollution as its greatest strength. However, hydrogen production and storage can result in relatively high costs. On the contrary, the cost associated with BEVs is the lowest among the NEVs and has a simple structure. However, the battery, which is the most important structure in BEVs, can encounter problems regarding its health status and aging. This makes the evaluation of the battery's SOH a vital research hotspot. Once this problem is solved, the BEVs can be able to replace ordinary cars entirely and become a significant turning point in social development.

3.2. Types of power battery in BEVs

The power battery is the primary power source in BEVs. It has two categories: storage battery and fuel battery. Fuel battery can generate electricity by converting chemical energy, using fuel and oxidant, into electric energy. The storage battery can be divided into the lead-acid battery, the lithium-ion battery (LIB), the nickel-hydrogen battery, and the sodium-sulfur battery (Zheng, 2016), and is suitable for BEVs. Having different performance and working principles, these battery types have certain advantages and disadvantages, which are summarized in Table 2.

As shown in Table 2, LIB is a popular choice because of its compact size, long life cycle, and has no associated pollution. It is estimated that by 2025, the global LIB market would exceed 100 billion dollars (Li et al., 2019a). Therefore, this article will be focusing the review on SOH prediction based on the LIBs.

3.3. Battery aging

LIB aging is the leading cause of change in battery health and life. The internal chemical reaction of the battery is hugely complicated. While explaining the cause of battery aging is not straightforward, a number of studies have been conducted in recent years that have tried to examine and explain the dynamics of battery aging (Barre et al., 2013a; Matsuda et al., 2019; Pelletier et al., 2017).

Based on the current literature, battery aging is mainly caused by external environmental factors and internal factors. External environmental factors refer to the location where the battery is situated and its operating environment, such as temperature (Wang et al., 2018a; Ma et al., 2014), charge and discharge rate (Gao et al., 2017a; Maher and Yazami, 2013), depth of discharge (DOD) (Meng et al., 2016; Omar et al., 2014), and the cut-off voltage of charging (Notten and Danilov, 2014; Gao et al., 2017b). The internal factors mainly refer to three influencing mechanisms: the loss of lithium inventory (LLI) (Jiang et al., 2019a; Castro et al., 2012), the loss of active material (LAM), and conductivity loss (CL). The LLI includes the generation of an SEI layer (Kim et al., 2013), the formation of lithium dendrites (Shen et al., 2017), and the self-discharge of a battery (Lang et al., 2016; Liu et al., 2012; Sarasketa-Zabala et al., 2015). The LAM comprises the decomposition of cathode material (Liu et al., 2012), the decomposition of anode material (Kim et al., 2013), and the decomposition of the electrolyte (Kassema et al., 2012; Li et al., 2015). The CL mainly refers to the aging mechanism that causes the battery's current collector to break and decompose and the battery adhesive to peel

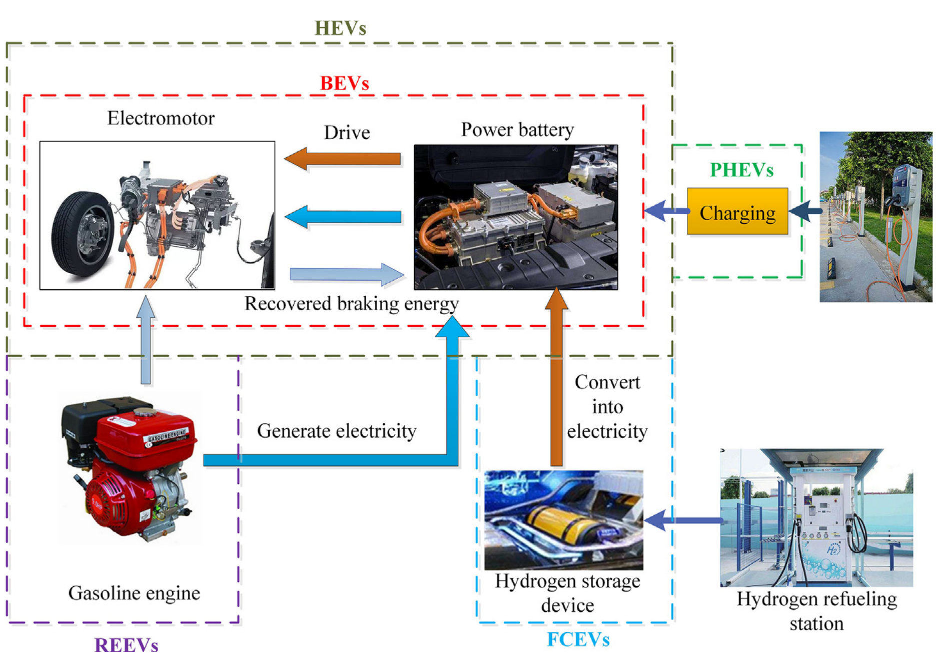


Fig. 3. The comparison of different types of NEVs.

Table 2
Types of power battery and their characteristics.

Type of power battery	Advantage	Disadvantage
Lead-acid battery	High technology maturity, low price, and high cost	High quality, low specific capacity rate, serious pollution.
Lithium-ion battery	Small volume, long cycle life, repeated charge and discharge	Life and performance are susceptible to external environments
Nickel-hydrogen battery	Compared with a lead-acid battery, it has greatly improved in terms of specific power, mass, volume, and density.	High-temperature requirements and high raw material prices
Sodium-sulfur battery	High specific energy, high power, fast charging	High operating temperature, high price, and poor safety
Fuel cell	High specific energy, environmental protection, and no pollution.	Complex system and lag in hydrogen supply system construction

off and degrade (Pastor-Fernandez et al., 2017; Yang and Dai, 2019). Fig. 4 presents a summary of reasons causing the aging of batteries.

4. SOH and RUL

Aimed at prolonging battery life, SOH and RUL prediction has developed into an important research hotspot. Different references consider them as either being identical or two completely different variables. Aside from the differences, similarities, and connections between the two that have sometimes been overlooked, some have misconstrued notions or inaccurate definitions for these terms. In order to clarify the discussion, this section will provide the working definitions for the terms, and examine the relationship between them.

4.1. Definition of SOH

The range describing battery health status is too broad that any factor that characterizes battery health can be used to define SOH. Most of the current research (Yao et al., 2018; Liu et al., 2019; Wang et al., 2018b) uses characterization parameters of battery aging to define SOH, such as capacitance, internal resistance, and power. But this approach can significantly distort the actual notion of battery health. Characterization parameters are not the only determinants of battery health, given that some of the battery's internal parameters also comprise the health category. So, it is necessary to establish a complete framework for defining SOH.

This section develops a broadly defined SOH, which includes both the failure and the degradation of batteries. Battery failure refers to parameters that are whether the voltage, current, temperature, battery SOC, self-discharge rate exceeds a specific normal range. Deterioration of the battery has been mentioned in the previous paragraph.

In a way, state of function (SOF) is more accurate in assessing battery health. SOF is assessed using battery factors SOC, SOH, recent usage, and current ambient temperature. Determining how long and how many times the battery works can be used to establish the SOF. However, there have only been a few studies on SOF, and there is currently no reliable model to correlate SOC, SOH, operating temperature, and the SOF (Shen et al., 2018).

4.2. Definition of RUL

The definition of RUL is fairly clear and straightforward. It indicates the period from when the observation is made to the

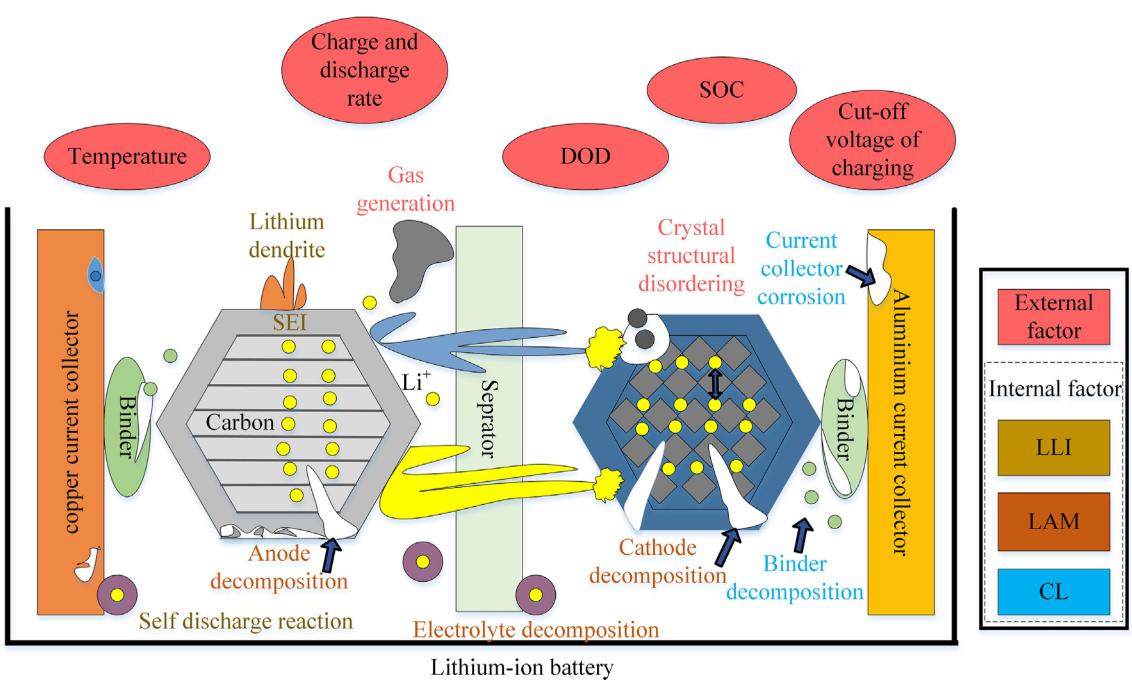


Fig. 4. Schematic diagram of the cause on battery aging.

estimated end of life (EOL). The EOL refers to the time and the number of charge and discharge cycles when the battery characterization parameter would reach the replacement threshold. The characterization formula of the RUL is as follows:

$$RUL_{\alpha} = \beta - \alpha \tag{1}$$

where α is the current cycle, and β is the cycle at the EOL. By predicting the EOL cycle number β, we can get the battery RUL at the α cycle.

4.3. Relationship between SOH and RUL

As previously mentioned, SOH refers to the ratio of the used LIBs' characterization parameters (e.g., battery capacity) to that of which when they are not used, which is given in percentage. RUL refers to the number of available cycles from the characterization parameters (e.g., battery capacity) of LIBs to the failure threshold, which is an integer value. Their definitions are shown in Fig. 5.

Based on their definition, there is a strong connection between SOH and RUL, since the aging parameter can be used to define both terms. Consequently, the SOH and the RUL can be estimated using the same methods from this aspect.

Currently, many references utilize the capacity as a parameter to evaluate the SOH and assess the RUL while predicting the SOH. In addition, several studies focus only on evaluating RUL by predicting the capacity. But according to the definitions described earlier, these methods can also be used in predicting SOH since capacity is also its characterization parameter. For example, a multi-scale EKF has been proposed to execute a joint estimation of SOC and SOH with dual time scales (Ma et al., 2019). The SOH is evaluated first by predicting the battery capacity, and then the RUL is estimated by making the SOH value as a characterization parameter by using Gauss–Hermite particle filter (GHPF). The block diagram of the joint estimation of SOC and SOH and the RUL prediction is shown in Fig. 6.

Literature (Wei et al., 2018) evaluates SOH and RUL simultaneously by predicting the capacity. The SOH and RUL estimation result is shown in Fig. 7.

In addition, the literature (Qu et al., 2019a; Zhang, 2018) use both battery capacity to predict SOH and RUL. A review article (Ungurean et al., 2017) even shows that RUL is directly related to SOH and introduced battery SOH/RUL estimation methods. Therefore, it can be seen from the above literature that the relationship between SOH and RUL is very close, and we can evaluate SOH and RUL at the same time by the characteristics such as capacity. So, many RUL prediction methods are also applicable to SOH estimation. In the next section, we will discuss the research status on SOH prediction combined with some RUL prediction methods.

5. Research status of SOH prediction

In Section 4, we have discussed the definition of SOH and RUL in detail and explained the possible sources of confusion in the current research, which is a good illustration of the difference and contact between them. At the same time, since RUL and SOH can be defined with the same characteristic parameters, some current RUL prediction methods can also be applicable for SOH estimation. Thus, in this section, we will discuss the current SOH prediction research status in detail, and combine some RUL prediction methods to make some supplements.

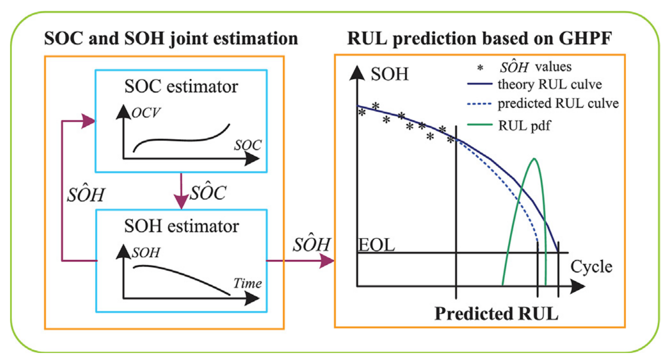


Fig. 6. Block diagram of SOC and SOH joint estimation and RUL prediction in Literature (Ma et al., 2019).

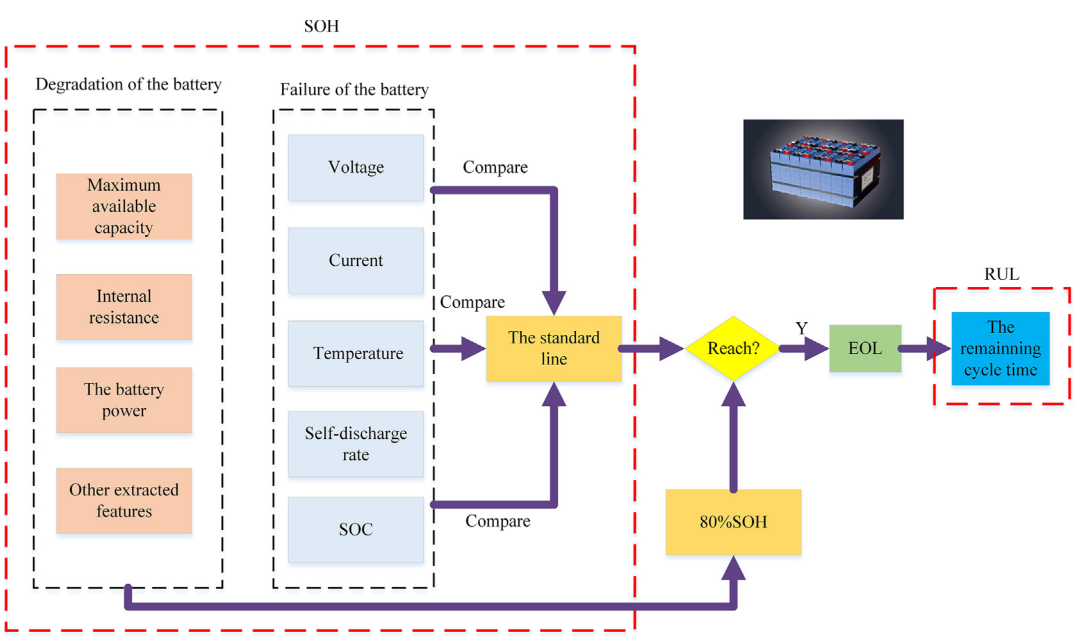


Fig. 5. The definition and relationship of SOH and RUL.

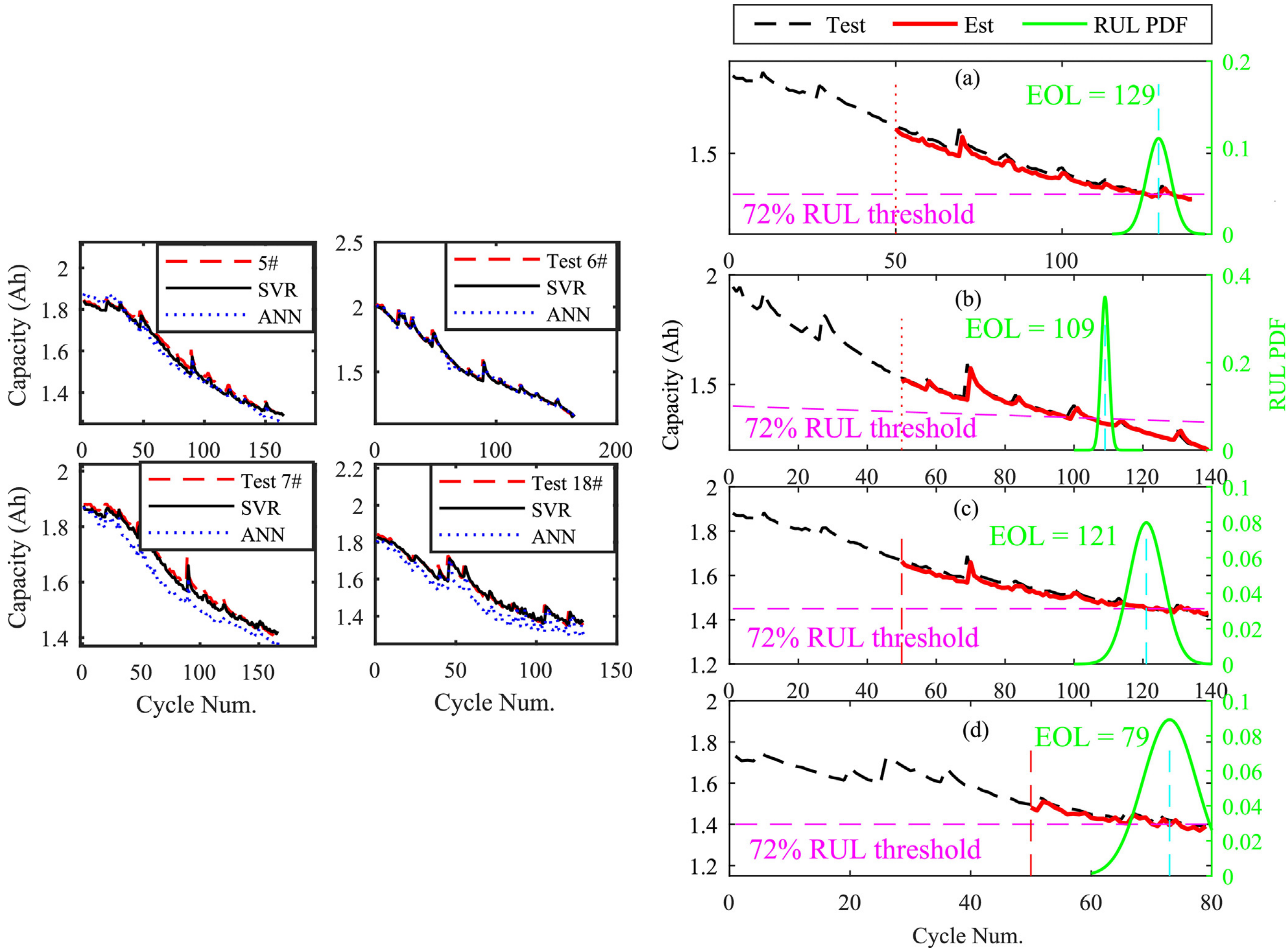


Fig. 7. The SOH and RUL prediction in Literature (Wei et al., 2018).

5.1. *Classification of SOH prediction methods*

There are numerous SOH prediction methods currently available. Many researchers have developed different classification systems for these prediction methods from different angles. These various classification systems could create some problems such as inaccuracy and confusion. Table 3 presents some classification systems found in the current literature.

According to the descriptions shown in Table 3, the classification standards vary for the different systems, but some commonalities can also be found. Most of the classification systems have model-based prediction methods, which has become the mainstream approach. Some references have identified other approaches, such as data-driven methods or random techniques, statistical methods, and adaptive methods. These methods are all based on data, which means that no matter how many subcategories are created, they all fall under the data-driven category. Some references consider IC, DV, and direct evaluation methods. Since these methods are neither from the perspective of a model nor are they data-driven, these methods are categorized together as ‘other methods’. For methods that combine the same or different types of processes, they are classified under the hybrid category.

Table 3
The classification of SOH prediction methods.

Literature	The classification of SOH prediction methods
Literature (Ungurean et al., 2017)	Coulomb counting method, Open Circuit Voltage (OCV) method, impedance spectrum method, and Kalman filtering method
Literature (Lipu et al., 2018)	Direct evaluation method, Adaptive method, Data-driven method, and Other methods
Literature (Barre et al., 2013a)	Electrochemical models, ECMs, Performance-based models, Empirical models, and Statistical methods
Literature (Barre et al., 2013b)	Model-based method: physical model and the ECM: Data-driven method: intelligent method and autoregressive prediction methods Hybrid methods.
Literature (Dai et al., 2019)	Direct measurement, Model-based methods, and Data-driven methods
Literature (Qian et al., 2019)	Be divided by utilizing characterizing SOH parameters: capacity, Direct current resistance, Alternating current resistance, incremental capacity (IC), and differential voltage (DV)

Based on the classification description and analysis of the previous SOH prediction techniques, these methods can be divided into four categories: model-based methods, data-driven methods, hybrid methods, and other methods. These four categories provide a complete picture of the current research status. Thus, we can describe the current SOH prediction method in this classification framework in order to make the discussion clear and organized.

5.2. Prediction method of SOH

As can be seen from the previous section, the current SOH prediction methods are divided into four categories: model-based methods, data-driven methods, hybrid methods, and other methods. These four categories provide a good overview of current SOH prediction studies. The following will be introduced respectively:

5.2.1. Model-based method

The model-based SOH prediction methods can be divided from different angles. At present, different literatures (Yang et al., 2017; Sun and Shu, 2011) have different classification criteria for model-based SOH prediction methods. For example: ECM, electrochemical model, mathematical model, life cycle model, physical model, filtering model, mechanism model, an empirical model and so on. We summarize the model-based SOH prediction method, they are classified into three categories: ECM-based method, electrochemical model-based method and mathematical model-based method.

5.2.1.1. ECM-based method. The ECM is a model that does not consider the chemical composition inside the battery and corresponding reaction. According to the electrical characteristics of the battery, the basic electronic components and the controlled voltage source are used to construct a model. At present, the commonly used ECM mainly included: Rint model, Thevenin model, PNGV model, first-order RC model, second-order RC model and composite model based on the above circuit improvement (Jiang et al., 2019b). The ECM instead of the polarization reaction and self-discharge reaction in the charging and discharging process by using resistors, capacitors, etc., so that the polarization effect and the reaction process are close to the actual situation of the battery. Therefore, the more the consideration, the more complicated the battery model, but the more the battery is in line with the actual use.

The basic ECM-based prediction method is:

① Select the appropriate ECM;
② Design the experimental scheme to identify the ECM parameters;
③ The internal resistance and the maximum SOC are used as state variables, and the indirect reaction SOH is solved by EKF or the like.

For the parameter identification for ECM, the most widely used experiment is the hybrid pulse power characteristic (HPPC) test. The parameter identification method is summarized here into two categories: Curve method of fitting comparison and Combining the complex frequency domain output equation of the circuit model with the least-squares (LS) method and so on. In the curve method of fitting comparison, the direct current (DC) internal resistance can be directly obtained from the sudden change of the voltage caused by the applied pulse current. For the polarization capacitance and polarization resistance solution, we should establish the time-domain voltage equation firstly and then compare the established time domain equation with a fitted voltage curve equation after the end of pulse charge and discharge, and then the parameters are obtained. Among them, the commonly used methods are: MATLAB curve fitting toolbox, optimization method (Banguero et al., 2018) and so on.

The circuit equation established in the first method is an equation of time domain, and the parameters can be obtained by comparing the fitted curve with the equation of establishing. So, it is simple and the result is not accurate, belongs to the offline parameter identification method. In addition, there is a classic parameter identification idea which establishes circuit equations in the complex frequency domain, and then using LS method (of course, there also has partial least square (PLS) and recursion least square (RLS)), maximum likelihood method to identify parameters online, these methods continuously modify the model parameters through the error between the model output and the actual output, and finally get the optimal model. The above-mentioned LS, PLS, RLS and other parameter identification methods can be used for online identification, the RLS and the improvement of RLS method are the most classic online parameter identification method. In literature (Kim et al., 2014), the authors propose a novel parameter identification method for model-based condition monitoring of LIBs. A fast UD factorization-based recursive least square (FUDRLS) algorithm is developed for identifying time-varying electrical parameters of a battery model, and the proposed method is more numerically stable than conventional parameter estimation methods based on the RLS.

All the above is the parameter identification methods of the ECM. Next, the identified parameters should be used to estimate the SOH. On account of the internal resistance and capacity can characterize the battery health effectively, and their changes reflect indirectly the changes in the battery health status, therefore we can use the established ECM combined with the filtering algorithm (extended Kalman filter (EKF), Dual-EKF (DEKF), and unscented Kalman filtering (UKF)) to estimate the battery ohmic internal resistance, which employs the ohmic internal resistance as the model of the state equation, the voltage as the model of the observation equation. For example, Literature (Zhou et al., 2019) studies an online in-use EV SOH estimation method using iterated Gaussian Process Regression- Extended Kalman filter (GPR-EKF) to incorporate LIBs data at the macro time scale and the micro time scale based on daily charge data of BEVs. In addition, fragment charge data at the micro time scale is adjusted with real-time iteration to be used as the state equation. So, the state and observation equations established in this literature are as follows in Equation (2) and Equation (3):

$$U_n^k = GP_k^f\left(U_1^k, U_2^k, \ldots U_{n-1}^k\right) + W_{n-1}^k \tag{2}$$

$$t_n^k = GP^h\left(U_n^k\right) + V_n^k \tag{3}$$

where n is total number of sampling time points when the battery reaches the cut off voltage V in the charge with constant current I, U_n^k is the measured charge voltage value at the charge moment, t_n^k is the charge moment. GP^h is a nonlinear measurement equation function based on GPR, GP_k^f is gained as the first state equation function of GP_0^f according to the first GPR of the full-charge curve.

Finally, the absolute moment corresponding to the initial voltage U, required full charge time for the kth charge t_k, and available capacity for the kth constant current charge are gained as following in Equation (4):

$$C_k = t_k \times I \tag{4}$$

where I is the current value of constant current charge of the battery.

5.2.1.2. Electrochemical model-based method. The electrochemical phenomenon inside the LIBs is very complicated, Li^+ is constantly embedded and disembedded, the SEI is continuously generated and changed, which causes the internal mechanism of the battery to change. The electrochemical model is based on various aging mechanisms. Among them, the model of the aging mechanisms has a mechanism model based on SEI and corresponding improved model, single factor electrochemical mechanism model and complex electrochemical mechanism model based on multiple comprehensive factors. It describes the microscopic physical and chemical processes inside the battery from the principle point of view quantitatively. Therefore, the aging mechanism model is an important class of electrochemical models.

There is also a class of EIS-based methods, which takes the battery as a circuit model consisting of a resistor, capacitor, and inductance (Liu et al., 2015). This method is to measure the impedance of the sine wave and then analyzes the electrode process dynamics, electric double layer and diffusion, so this method also belongs to the category of methods based on electrochemical models. From the above analysis, in this section, the electrochemical model-based method is divided into: the aging mechanism-based method and the EIS-based method.

(1). The aging mechanism-based method

The aging mechanism-based method describes the battery dynamic parameters, the thermodynamic parameters, and the electrical properties of the material from the perspective of the internal physicochemical process and aging. Therefore, we can model the electrochemical reaction of Li^+ concentration, SEI thickness, and electrode conductivity during battery aging and establish the equation to study SOH (Sankarasubramanian and Krishnamurthy, 2012; Waag et al., 2013).

Githin et al. (Prasad and Rahn, 2013), propose and simplify an electrochemical model, and then obtain the impedance transfer function according for a third-order Padé approximation, characterize SOH by two key aging parameters: cell resistance and the solid phase diffusion time of Li^+ species in the positive electrode, which can be expressed as following in Equation (5) and Equation (6):

$$\hat{S}\,OH_{R_T} = \frac{\hat{R}_T(t)}{\hat{R}_T(0)} \tag{5}$$

$$\hat{S}\,OH_{\tau_D} = \frac{\hat{\tau}_D(t)}{\hat{\tau}_D(0)} \tag{6}$$

where the $\hat{R}_T(t)$ represents the current cell resistance, $\hat{R}_T(0)$ is the initial resistance, $\hat{\tau}_D(t)$ is the current solid phase diffusion time of Li^+ species in the positive electrode.

The literature (Safari and Delacourt, 2011) establishes an electrochemical model simulating the growth of SEI membranes, this model reveals the effect of SEI membranes on battery capacity degradation. Otherwise, an improved single-particle model (SPM) is also developed to evaluate the capacity decay mechanism of a lithium iron phosphate battery understanding and cycling conditions. In literature (Li et al., 2018a), an SP-based degradation model is developed by including SEI layer formation. The initial battery capacity of a fresh cell is estimated to be as following in Equation (7):

$$Q_{in} = q_r Q_g \left(\frac{4}{3}\pi r_n^2 \rho_g\right) \tag{7}$$

where Q_g is the specific capacity of graphite, ρ_g is the density of graphite, and q_r is a formation cycle efficiency, which is assumed to be 0.9. The battery capacity after the SEI formation cycle is:

$$Q_0 = 0.9 Q_{in} \tag{8}$$

Then, the capacity fade rate per cycle after the formation cycle is:

$$\frac{dQ_N}{dN} = -\frac{n_{SEI}\rho_{SEI}F}{M_{SEI}}\frac{dV_{SEI}}{dN} \tag{9}$$

where n_{SEI} is the consumed Li^+ for 1 mol of SEI layer formation, ρ_{SEI} is the density of SEI films, F is the Faraday's constant, M_{SEI} is the molecular weight of compounds constituting SEI, N is the cycle number.

(2). The EIS-based method

EIS is an important technical means for the internal reaction principle and the battery aging. The simplest EIS-based method is to measure the alternating current (AC) impedance of the battery to evaluate the SOH. In literature (Xia and Abu Qahouq, 2019), the relationship between the AC complex impedance phase and its capacity fading due to aging is utilized to estimate the SOH. It finds that the zero-crossing frequency of battery impedance phase can reflect the aging status, which uses the zero-crossing frequency of battery impedance phase as input of ANN to predict SOH.

5.2.1.3. Mathematical model-based method. The mathematical model contains a wide range. It is reasonable to say that any model with mathematical formulas or other mathematical forms should belong to the mathematical model, but this understanding is too absolute. The current mathematical model-based methods have empirical models and statistical models. But the statistical model tracks the battery degradation information based on the statistical random filtering algorithm generally, these methods are a complete system and they rely on data particularly. So, filtering algorithms are classified into methods based on data-driven models.

In addition, some people also mentioned the probabilistic model-based method. The probabilistic model-based method is derived from the probability theory and belongs to a mathematical model. Therefore, this paper divides the methods based on mathematical model into empirical degradation model-based method and probability model-based method.

(1). Empirical degradation model-based method

The basic empirical degradation model-based method is to model the relationship between battery life attenuation and temperature, charge-discharge ratio, and over-charge and over-discharge of the battery. The model is tested under different attenuation factors, and vast data are fitted to a SOH expression with the attenuation factor as a variable. Therefore, the more attenuation factors are used, the more accurate the SOH prediction is.

In literature (Singh et al., 2019), the semi-empirical model used is described by Equation (10), the extracted parameters were used to calculate the SOH over the entire battery life.

$$SOH = 1 - \left(\frac{1}{2}k_1 N^2 + k_2 N\right) - \frac{k_3}{Q_{\max}(fresh)} i \tag{10}$$

where k_1, k_2, and k_3 are the coefficients that depend on battery operation conditions. N represents the number of charge-discharge cycles the battery experienced at the time of this SOH calculation, and i is the discharging current.

Sebastian Paul (Paul et al., 2013) and others point out that the battery will have a capacity recession in the static environment, the ambient temperature, the hold time and the SOC can affect the battery life, and supplement the basic calendar aging and cycle aging experience formulas. The calendar aging and cycle aging experience formulas is shown in Equation (11) and Equation (12):

$$C_{loss,cal} = A \cdot \exp\left(\frac{-E_A}{R \cdot T}\right) \cdot t^B \cdot SOC^C \tag{11}$$

$$C_{loss,cycl} = f(SOC, T, C) \cdot EN^D \tag{12}$$

where the $A \cdot \exp\left(\frac{-E_A}{R \cdot T}\right) \cdot t^B$ is Arrhenius formula, A, B and C are the coefficient to be fitted. $f(SOC, T, C)$ is a concentrated function of the effects of various stresses on the rate of capacity loss during the cycle, this article only selects the SOC, T, and discharging rate. EN is the total circulating power throughput.

Besides, John (John et al., 2011) establishes a power battery capacity attenuation model with temperature and discharge rate double-life decay acceleration stress based on the research of Bloom, and the accuracy of model prediction reached more than 80%. The empirical formula they proposed is shown in Equation (13) and Equation (14):

$$Q_{loss} = B \cdot \exp\left[\frac{-31700 + 370.3 \cdot C\_rate}{RT}\right] \cdot {A_h}^{0.55} \tag{13}$$

$$A_h = 2 \cdot n \cdot DOD \tag{14}$$

where R is the molar gas constant, T is the cycle temperature, n is the cycle number, C_rate is the discharging rate and DOD is the discharging depth.

Su (Su et al., 2016) studies systematically seven major factors affecting battery degradation using orthogonal experimental design, and he introduces an empirical aging model and a deceleration factor for comparing the decay characteristics under different test conditions.

(2). Probability model-based method

Another battery model that can perform SOH estimation is the probability model, sometimes called the probability density function (PDF) method. Its basic idea is to calculate the occurred probability of different voltage value points in the voltage detection data, when in the charging or discharging curve of the battery for different states of aging, finally drawing a probability density curve. With the battery aging, the corresponding voltage of the peak will change, so the mapping between the peak voltage of the probability density curve and the SOH can be established (Li et al., 2014a).

In literature (Feng et al., 2013), Feng proposes a method for estimating SOH based on PDF, this method originates from probability theory and estimates SOH by analyzing the charge-discharge voltage curve in BEVs. The most basic method can be described by the following formula:

$$SOH = \frac{N_{cyc}}{N_{fresh}} \tag{15}$$

where the N_{cyc} is the frequency of the voltage within 3.38V−3.42V during charging (discharging) when the LIBs is used and the N_{fresh} is the number of times when the battery is fresh.

5.2.2. Data-driven method

Data-driven is a method that builds a rough model and then refines the data with vast data to make the model consistent with the data. For complex electrochemical dynamic systems in LIBs, the model-based methods are often complex and difficult to implement, but the data-driven method does not consider the electrochemical reaction and the failure mechanism inside the LIBs. So, the data-driven prediction methods become a research hotspot. This section refers to the methods used in many literatures according to the principle and applicable conditions of the methods used, the data-driven SOH prediction methods are divided into artificial intelligence-based method, filtering-based method, statistical data-driven method, time series-based method.

5.2.2.1. Artificial intelligence-based method. The artificial intelligence-based method establishes the mapping relationship between characteristic parameters and lifetime of LIBs degradation through an artificial neural network (ANN), support vector machine (SVM) and Relevance Vector Machine (RVM) and other intelligent algorithms such as the machine theory and grey theory and fuzzy algorithm, and then these methods extrapolate the estimated SOH and RUL. Therefore, the artificial intelligence-based methods combining the current research status are divided into ANN, SVM, RVM, fuzzy algorithm, and integrated learning algorithm.

(1). ANN

ANN has been widely used in self-organization and self-learning, and it does not rely on the electrochemical principle inside the battery. Yang et al. (Yang et al., 2016) uses maximum available capacity to indicate the SOH based on a back propagation (BP) neural network, He takes the parameters of the first-order ECM and SOC as the input of BP and SOH as the output, and trains the BP to predict SOH. The literature (Pan et al., 2018) finds that characteristics like voltage of the platform, duration of platform, voltage boost points and length of the curve are all diverse among batteries under different cycle numbers. Thus, battery terminal voltages during constant current charging subprocess are set as the inputs of the FFNN and battery equivalent circle life is the output. This method can predict SOH by changing the output of model.

Besides, many researchers have improved on ANN and propose many other types of neural networks. For example, Rezvani, et al. (Rezvain et al., 2011) estimate the battery SOH given a time series of capacity over cycles, it the number of inputs to the adaptive neural networks (AdNN) is n, the training portion of the dataset is the capacity data of the last n cycles considered as an input and based on the relation of those capacities the model is going to estimate the capacity of cycle n+1, which achieves one step-ahead SOH prediction results using AdNN. The SOH prediction method with AdNN in this literature is shown in the following Fig. 8:

(2). SVM

SVM is a supervised machine learning method proposed by Vapnik in 1995. In response to the prediction problem of SOH, Adnan Nuhic et al. (Nuhic et al., 2013) point out that the SOH is

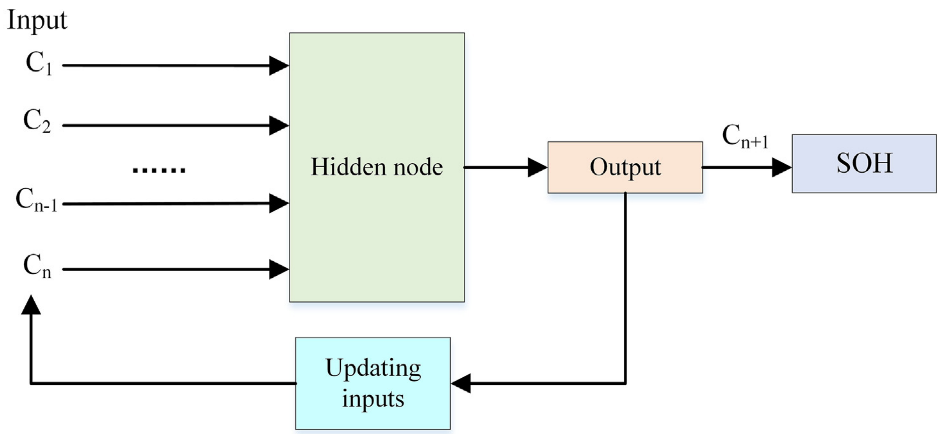


Fig. 8. The SOH prediction method with AdNN.

related to the actual capacity, and they use the SVM method to estimate the SOH and remaining service life from the perspective of capacity. The training data comes from the first 11 cycles of charge-discharge test and the last seven cycles are predicted by regression. The results show that SVM can predict the available capacity accurately and estimates the SOH and RUL effectively.

In addition to the basic SVM algorithm, there are many studies on improving the SVM algorithm and the fusion of the SVM algorithm with other algorithms. Pattipati et al. (Pattipati et al., 2011) introduce the Hidden Markov model (HMM) into SVM, and they realize the battery RUL prediction. Among them, they exploit the high degree linear correlation between the battery resistance and capacity, and the nonlinear SVM models are used to forecast the capacity fade. In literature (Chen et al., 2018a), a fixed size LS-SVM model that is based on the arbitrary entropy is constructed to estimate the SOH. Firstly, the authors select the voltage range and then use the discharge time of the voltage interval as the input variable of the model, SOH as output variable. Finally, the Bayesian framework is used to estimate the relevant parameters in LS-SVM, which increases greatly the calculation speed.

(3). RVM

In recent years, the RVM model has been gradually used for the prediction, but the application is relatively small. In the literature (Qin et al., 2017), the authors use grey relation analysis (GRA) to select features. Firstly, the duration of equal discharging voltage difference (DEDVD) and the duration of equal charging voltage difference (DECVD) are selected as the input of the model and then followed by feature vector selection (FVS) to remove the redundant points in the input data. Finally, the RUL is predicted using RVM. Since battery capacity is used as the output of the model, it can also be used to predict SOH. In literature (Li et al., 2014b), a mean entropy-based method is proposed to select the optimal embedding dimension for correct time series reconstruction. Firstly, it selects the optimal embedding dimension, then constructs the input variables for the RVM prediction model by the optimal embedding dimension, which predicts the future SOH. Among them, the pattern of training data set is as shown in Table 4.

Table 4
Pattern of training data set.

i	x_i				t_i
1	X_1	X_2	…	X_m	X_{m+1}
2	X2	X3	…	X_{m+1}	X_{m+2}
	…	…	…	…	…
n-m	X_{n-m}	X_{n-m+1}	…	X_{n-1}	x_n

Where the n is the amount of training data set, m is the optimal embedding dimension.

(4). **Fuzzy logic method**

The fuzzy logic is a behavior-based bionic reasoning method and is mainly used to solve complex reasoning problems with fuzzy phenomena. In literature (Chen et al., 2019a), the authors use the cycle number, voltage drop value and internal resistance change as the input quantity, and the weight of the corresponding SOH as the output. Among them, the T-S fuzzy control is used to establish the dynamic prediction model, and the SOH is calculated according to the weight and boundary conditions. In literature (Kim, 2014), the fuzzy logic is used to predict SOH, where the input is the maximum capacity and resistance. Among them, the maximum capacity is defined by the DOD and is calculated according to the OCV and SOC, the resistance is defined by the ratio of the OCV to the current-voltage and the current, the effect of temperature on the resistance is also considered. After determining these two variables, the membership function is selected according to the established fuzzy rules to predict the SOH.

(5). Integrated learning

Integrated learning is a data-driven method based on machine learning and uses a series of learners to learn, finally, using some rules to integrate the learning results to achieve better learning effect than a single learner. Currently, the integrated learning algorithms which used commonly are Boosting, Bagging, random forest (RF), and gradient boosting decision tree (GBDT).

At present, there are currently two integrated learning methods based on the SOH prediction. One is the AdaBoost algorithm, the AdaBoost algorithm combines the performance of the weak learning machine in the learning and training process. The literature (Li et al., 2019b) starts from the parameters that can characterize SOH and adds the concept of sample entropy and selects six variables as the input of the model. Finally, the model prediction is carried out by Adaboost.RT.

Another method is to combine the machine of weak learning with the integrated learning theory. In literature (Chen et al., 2018b), the authors combine the SVM algorithm with of integrated learning theory, and they propose a way with ensemble SVM based on local information fusion. The algorithm divides the original training set into several sub-training sets and integrates the trained multiple SVR models with the integrated learning algorithm.

5.2.2.2. Filtering-based method. The filtering-based method is one of the most used methods in robust estimation. The calculation of the filtering method usually involves two steps of prediction and correction. We have already mentioned this kind of filtering technique based on the ECM in section 5.2.1, it establishes a state-space model by constructing a state equation and uses parameters (internal resistance, capacity, etc.) that characterize SOH as state variables, thereby dynamically tracking and predicting the health, and then using the filtering algorithm for iterative solution. So, the common algorithms based on filtering include KF, PF and improved algorithm based on KF (PF, etc.). They can obtain the variation of test data and time from the data point of view and the internal recursive relationship of the system state.

(1) KF

The KF is an algorithm that uses the linear system state equation to estimate the system state through the input and system output. Since the observed data includes the effects of noise and interference in the system, the optimal estimate can also be considered as a filtering process. However, the traditional KF algorithm is a linear system-based filtering algorithm, and the battery system is a complex nonlinear system. Therefore, many studies have proposed the improved algorithms such as EKF, DEKF, and UKF.

1). EKF

The EKF is like the classical linear Kalman algorithm. The equation of state and the measurement equation are shown in Equation (16):

$$\begin{aligned} x_{k+1} &= f(x_k, u_k) + w_k \\ y_k &= g(x_k, u_k) + v_k \end{aligned} \tag{16}$$

where $f(x_k, u_k)$ is a nonlinear function related to the state of the system, and $g(x_k, u_k)$ is a nonlinear function associated with both state variables and observed variables.

In literature (Cheng et al., 2008), the authors use EKF to estimate the capacity and internal resistance. The real-time estimation of the SOC makes these two characteristic parameters updated in real-time and provides the necessary parameter information for SOH prediction. The state space model with ohmic internal resistance as state variable as shown in Equation (17).

$$\begin{cases} R_{0,k+1} = R_{0,k} + r_{1,k} \\ U_{t,k} = V(SOC_k) - U_{1,k} - U_{2,k} - R_{0,k}I_{t,k} + \varepsilon_{1,k} \end{cases} \tag{17}$$

where $r_{1,k}$ is the process noise of the system, and $\varepsilon_{1,k}$ is the interference noise in the system output variable. There is a SOC value in the observation equation of the internal resistance estimation, so the internal resistance prediction is completed by the estimated value of the SOC previous time, and the SOC can be obtained by ampere-hour integration method.

In addition to, in order to reduce the computation cost and enable deployment of the BMS on the low-cost hardware, a Lebesgue-sampling-based EKF (LS-EKF) is developed to estimate the SOH and SOC, and the LS-EKF can eliminate unnecessary computations, especially when the states change slowly (Yan et al., 2019). The state and observation equations established in this literature are as follows in Equation (18):

$$\begin{cases} \hat{x}_{t_{k+1}} = f\left(\hat{x}_{t_k}, u_{t_k}, D_{t_k}\right) + w_{t_k} \\ z_{t_k} = h\left(x_{t_k}\right) + v_{t_k} \end{cases} \tag{18}$$

where f is a nonlinear function, u_{t_k} is the input, which includes environmental factors that affect battery capacity degradation, $\hat{x}_{t_k}$ is the SOH state, w_{t_k} is the Gaussian noise, D_{t_k} is the Lebesgue state length given by the distance between two adjacent Lebesgue states. z_{t_k} is the measurement, which is the battery capacity for the SOH estimation and terminal voltage for SOC estimation.

2). DEKF

The DEKF algorithm uses two EKF alternately, which is mainly divided into an iterative form of the DEKF algorithm and a sequential form of the DEKF algorithm. The iterative form of the DEKF algorithm is mostly used offline, and the state and parameter estimation are performed by the DEKF algorithm on the premise of acquiring all the data. The sequential form of the DEKF algorithm is to use the current measured values, update the model and system state, and finally, it performs real-time estimation.

In literature (Fang et al., 2019), based on the relationship between the ohmic internal resistance and the SOH, a joint estimator using the DEKF algorithm is proposed for the estimation of both SOC and SOH. Among them, according to the change of the ohmic resistance, the discrete state space equation of the internal resistance and the output observation equation of the battery model can be described as:

$$R_0(k) = R_0(k-1) + r(k-1) \tag{19}$$

$$U(k) = U_{oc}[SOC(k)] - U_{R_cC_c}(k) - U_{R_dC_d}(k) - i(k) \times R_0(k) + v(k) \tag{20}$$

where R_0 is the ohmic resistance of second-order ECM, $r(k-1)$ represents the noise of ohmic resistance, R_c, R_d and, C_c, C_d are polarizing capacitors and polarizing resistors, $v(k)$ represents the noise of output observation.

In literature (Guo et al., 2017), the authors use the DEKF algorithm to estimate SOC and ohmic resistance online, and then estimate the SOH of each cell in the battery pack online based on the relative state of the health assessment method. Finally, the lithium iron phosphate battery is tested under different conditions, and the accuracy and effectiveness of the method are verified. In literature (Kim et al., 2012), the authors present a complementary cooperation algorithm based on DEKF combined with pattern recognition, it uses for an application Hamming neural network to the identification of suitable battery model parameters, which avoids the need for repeated parameter measurement.

3). UKF

The UKF was proposed by Julier in 1969, the probability density Julie of the approximate nonlinear function is solved by UT transform. This method is better than the EKF method which makes the estimation error is reduced. The computational complexity of this UKF algorithm is also not increased. In literature (Chen et al., 2014), the authors estimate SOH through UKF, where UKF uses internal resistance and SOC as state parameters, and predicts indirectly SOH by estimating SOC while continuously adjusting internal resistance. Among them, this literature uses Rint model, the state and observation equations established in this literature are as follows in Equation (21) and Equation (22):

$$\begin{cases} SOC(k+1) = SOC(k) - \dfrac{I(k)\cdot\Delta T}{Q_{\max}} + w_1 \\ R(k+1) = R(k) + w_2 \end{cases} \tag{21}$$

$$V(k) = V_{ocv} - I(k)\cdot R(k) + C + \varepsilon \tag{22}$$

where $Q_{\max}$ is the rated capacity of the battery, w_1 and w_2 are the Process noise, ε is the measurement noise, C is the parameter that corrects the error.

In literature (Chen and Wang, 2014), the Ohmic resistance of the battery model is identified online based on the UKF and the estimation model of the SOH for the 18650-type battery is established. In addition to this, its ECM is that the inconsistent factor during the charging/discharging working periods is considered. The online-estimation equations based on the standard UKF are defined as follows:

$$R_0^{k+1} = R_0^k + r_k \tag{23}$$

$$U_k = F[SOC_k] - U_k^1 - U_k^2 - R_0 I_k + \nu_k \tag{24}$$

where r_k is the Ohmic resistance and its value changes slowly during the working period. U_k^1 and U_k^2 are the voltage at each end of the polarization resistor resistance. formula (24) describes the output measurement function based on the Ohmic resistance. ν_k is the estimated error of the battery SOH.

In literature (Wang et al., 2016), the SOH prediction method for accurately estimating the 18650 high-capacity LIBs are further studied based on UKF. The 500-cycle charge-discharge experiment is completed, the authors obtain the relationship of the internal resistance and SOC, the relationship of the internal resistance and cycle number. Finally, the SOH estimation model is established based on the UKF estimation method and verified by simulation.

(2) PF

PF is a statistical filtering method based on the Monte Carlo method and Bayesian estimation. It can deal with effectively complex nonlinear systems in practice and has become a developing direction of modern filtering theory.

In literature (Dong et al., 2018), the authors concern with online short-term SOH estimation and long-term RUL prediction using the Brownian motion (BM) based degradation model and PF. Firstly, the proposed model tackles the capacity degradation as the traveling distance of a Brownian particle in each time interval, and the BM-based battery degradation models as shown in Equation (25).

$$y_i = \lambda_{t_i} t_i + \sigma_B B_t + \nu_{t_i} \tag{25}$$

where y_i represents the percentage of the capacity loss in the BM-based battery degradation models, t_i represents cycle number, B_t is the standard BM representing the stochastic dynamics of degradation behaviors. ν_{t_i} denotes an additive Gaussian model error. λ_{t_i} is the drift parameter.

Then, the PF is used to estimate the drift parameter of the BM, and the state transition equation can thus be formulated as:

$$\hat{\lambda}_{t_i} = \hat{\lambda}_{t_{i-1}} + \eta_{t_{i-1}} \tag{26}$$

where $\hat{\lambda}_{t_i}$ and $\hat{\lambda}_{t_{i-1}}$ are estimated drift parameters at time t_i and t_{i-1}, $\eta_{t_{i-1}}$ is an additive Gaussian noise.

Dong et al. (Dong and Jin, 2014) establish the battery electrochemical impedance model. Among them, the time dependent aging of impedance can be modeled by an exponential function:

$$R = R_0 \exp(\lambda_R t) \tag{27}$$

where R represents R_e or R_{ct} which in the electrochemical impedance model, R_0 is a constant and λ_R is the impedance aging parameter which is estimated through the PF.

The state and observation equations established in this literature are as shown in Equation (28) and Equation (29):

$$\begin{cases} \lambda_{R,K} = \lambda_{R,K-1} + V_{R,1,K} \\ \widetilde{R_K} = \widetilde{R_{K-1}} \exp(\lambda_{R,K-1} \triangle K) + V_{R,2,K} \end{cases} \tag{28}$$

$$R_K = \widetilde{R_K} + \eta_{R,K} \tag{29}$$

where the impedance aging parameter is represented by $\lambda_{R,K}$. The smoothed impedance is represented by $\widetilde{R_K}$. The state noise is denoted by $V_{R,1,K}$ and $V_{R,2,K}$. The system measurement R_K represents impedance R_e or R_{ct}. The measurement noise is represented by $\eta_{R,K}$.

In literature (Zhang et al., 2014a), the PF algorithm is used to estimate the battery SOH of the, and the end of discharge (EOD) is selected as the prediction parameter.

5.2.2.3. Statistical data-driven approach. The statistical data-driven method is based on statistical theory and combines with other mathematical principles. Statistical data-driven methods generally have two categories: GPR and wiener process (WP).

(1). GPR

The GPR is a data-driven method proposed by Williams and Rasmussen. It is a new machine learning method based on Bayesian theory and statistical learning theory. The GPR is suitable for dealing with complex regression problems such as high dimensionality, small sample, and nonlinearity.

In literature (Peng et al., 2018), a prediction method fusing the wavelet de-noising (WD) method and the hybrid Gaussian process function regression (HGPFR) model for predicting the RUL is proposed, this method aims to overcome the problem that the trend fitting deteriorates rapidly when test data are far from the training data for multiple step ahead estimation. Because this literature uses battery capacity as a parameter to characterize RUL, it can predict SOH at the same time. Liu et al. (Liu et al., 2013) use GPR to predict the lifetime and represent the predicted SOH by mean and variance, which can describe the uncertainty in the prediction process, in addition to that, the linear Gaussian process function regression (LGPFR) and the GPFR with quadratic polynomial mean function (QGPFR) is designed to achieve the advance estimation and proves to have a good effect.

(2). WP

The WP is a typical stochastic process and belongs to the so-called independent incremental process, which starts from the theory and application of stochastic processes. WP (Liu et al., 2017) can describe not only the monotonic degradation of the device performance but also the non-monotonic degradation of the device.

According to the above description, they follow a certain law of statistical and can be studied through probability and statistics since the random processes are irregular. The literature (Xu et al., 2019) mainly studies the impact of relaxation effect on the degradation law and proposes a novel SOH estimation method based on

WP. Among them, the life cycle is divided into three parts. Firstly, the degradation model after eliminating the relaxation effect is established based on linear WP, which can be expressed by Equation (30):

$$X(t) = x_0 + \lambda t + \sigma_B B(t) \tag{30}$$

where x_0 is the initial capacity, λ is the drift coefficient, which characterizes the degradation rate, σ_B is the diffusion coefficient, and $B(t)$ is the standard Brownian motion representing the dynamic characteristics. So the SOH estimation of LIBs after eliminating the recovery process can be written as shown in Equation (31):

$$SOH_1(t|x_{1:k}) = \frac{x_k + \mu_1(t - t_k) + \sigma_{B_1} B(t - t_k)}{x_0} \tag{31}$$

where x_k the is the current cycle capacity, x_0 is the initial capacity, $x_{1:k}$ are the on-site degradation data at the time $t_1 \ldots t_k$.

The capacity regenerated model is developed by the normal distribution can be expressed as shown in Equation (32):

$$g(\Delta t^s) = a(\Delta t^s)^b \tag{32}$$

where the $g(\Delta t^s)$ is assumed that the mean of regenerated capacity after the rest time Δt^s. So the SOH estimation of LIBs after the degradation model of regenerated capacity can be written as shown in Equation (33):

$$SOH_2(t|x_{1:k}) = \frac{x_k + \sum_{i=k}^{t-1} \mu_3(t_{i+1} - t_i)}{x_0} \tag{33}$$

The degradation model of regenerated capacity is established based on a nonlinear Wiener process, which can be expressed by Equation (34):

$$Y(t) = y_0 + \eta \Lambda(t : \theta) + \sigma_{B_2} B(t) \tag{34}$$

where y_0 is the initial capacity, $\eta \Lambda(t : \theta)$ is a nonlinear drift coefficient, and σ_{B_2} is the diffusion coefficient.

5.2.2.4. Time series-based method. To put it simply, time series-based method predicts future development based on past trends. The most basic time series-based prediction methods are: simple sequential mean method, weighted sequential mean method, moving average method, weighted moving average method, and so on. These methods generally give different weights according to the degree of the data influence at different times in the same moving segment on the predicted value, and then the average value is used to predict future values. In addition to the most basic time series-based methods, Autoregressive moving average model (ARMA), long-short term memory (LSTM), and recurrent neural network (RNN) are also important methods for studying time series prediction. These methods are more complicated, but the effect is better, which are popular methods based on time series prediction currently. Among them, RNN and LSTM belong to the neural network, but these two methods have good prediction effects based on time series, so it is reasonable to place them in the time series-based method.

(1). ARMA

ARMA is an important method for studying time series. It is composed of an autoregressive model (AR model) and a moving average model (MA model). The biggest problem with these methods is that the AR order and the MA order are difficult to determine, so the current research combines many optimization algorithms.

Long et al. (Long et al., 2013) use the AR model to track the decay process of available capacity and use the PSO algorithm to determine the model order, eventually, they complete the online RUL estimation, which can also be used as a method to evaluate SOH. Of course, there are improved algorithms for ARMA such as autoregressive integrated moving average (ARIMA). In literature (Zhou and Huang, 2016), a novel approach that combines empirical mode decomposition (EMD) and ARIMA model is proposed for RUL prognostic in this paper. Firstly, the EMD is used for decomposition SOH time series, and then the decomposed sequence is predicted by ARIMA and finally combined, the basic prediction procedure is shown in Fig. 9. Among them, it takes the battery capacity as the parameter representing SOH, and then uses SOH to characterize RUL and predict, so it also applies to the SOH prediction.

(2). RNN

RNN is an ANN in which nodes are connected in a loop. The internal state of such a network can exhibit dynamic timing behavior. In literature (Eddahech et al., 2012), the authors establish the battery model and monitor the cell model parameters using EIS measurements, capacity and equivalent series resistance (ESR) values predicted using RNN are used as an indicator of SOH, and the SOH evaluation process as shown in Fig. 10.

(3). LSTM

LSTM is a time-cycle neural network specially designed to solve the long-term dependence of general RNN. In addition to, the LSTM adds input gates, forgetting gates and output gates compared to the RNN.

In literature (Qu et al., 2019b), Qu establishes a prediction model for SOH through LSTM, and predicts SOH based on a sliding window, the basic model formula is shown in Equation (35). At the same time, in order to make full use of the latest data to improve the accuracy of the model, an incremental learning mechanism is introduced in the realization of SOH monitoring, where SOH is defined by capacity.

$$SOH_{t+1}^p = f\left(\left[SOH_t^r, SOH_{t-1}^r, \ldots SOH_{t-w+1}^r\right]\right) \tag{35}$$

where SOH_{t+1}^p is the prediction value of step t, SOH_t^r is the

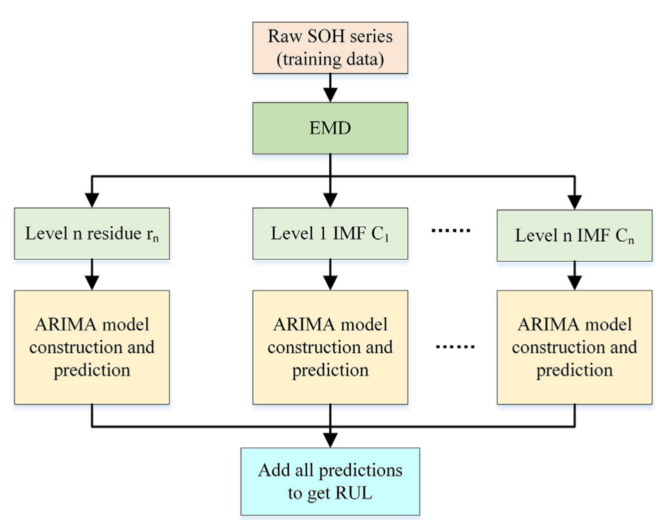


Fig. 9. The basic prediction procedure in literature (Zhou and Huang, 2016).

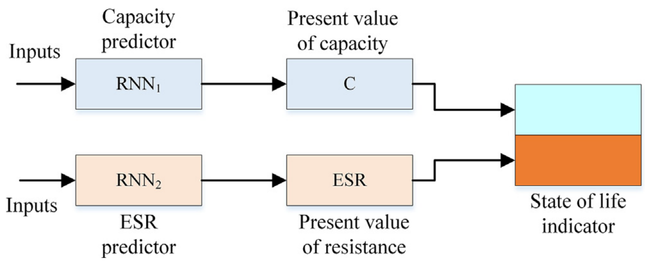


Fig. 10. The SOH evaluation process in literature (Eddahech et al., 2012).

observation value of step t, w is the length of slide window.

In literature (Chen et al., 2018c), a SOH estimation method using LSTM is applied to predict battery life for BEVs. Among them, the discharging time under constant current, the number of charging and discharging cycles, and the charging capacity are employed to build the prediction model with LSTM. The internal modeling parameters are trained by public battery datasets, in which the discharging process is introduced for battery SOH prediction.

5.2.3. Hybrid methods

As a name suggests, hybrid methods are a combination of many different methods. At present, the hybrid methods generally have a combination of the same type methods and a combination of different type methods. In addition, there are combinations of optimization algorithms and other methods, these methods generally achieve better performance by optimizing the model parameters and thresholds.

As to the combination of the different type SOH prediction methods, Zenati, et al. (Zenati et al., 2010) develop a hybrid method based on an EIS-based method and a fuzzy logic method. As they find differences between impedance of new and aged cells, so they conclude that using EIS measurements to evaluate the SOH by using fuzzy logic method. Among them, inputs of the fuzzy logic system (FLS) will be based on previous data extracted from EIS measurements. Zhao et al. (Zhao et al., 2019) propose a hybrid method based on an empirical degradation model-based method and a data-driven method. They extracts the SOH values of the regeneration cycle, the regeneration cycle number, and normal degradation from the SOH sequence and predict them for considering the effect of the regenerative phenomena. Among them, in the normal degradation prediction phase, they use the RVM and the grey model (GM) alternately to carry out a multi-step SOH prediction based on the time series. As for the capacity regeneration stage, using the exponential degradation model to model the regeneration region.

Liu et al. (Liu et al., 2014) propose a hybrid method based on an empirical degradation model-based method and a time series-based method. They find the degradation trend will be accelerated with this degradation process developed, so they denote the novel model as nonlinear degradation AR model (ND-AR model), and ND-AR model is defined as follows in Equation (36):

$$x_t = K_T \times \left[\Phi_1 x_{t-1} + \Phi_2 x_{t-2} + \ldots \Phi_p x_{t-p} + a_t\right] \tag{36}$$

where the K_T is the "accelerated" factor and the K_T as follows in Equation (37):

$$K_T = \frac{1}{1 + a \times (k + b)} \tag{37}$$

where k is the prediction step, a and b are the parameters that should be identified with the training data samples.

Then, the ND-AR model capacity prediction as observation of PF and update state transition equation, among them, the state transition model as shown in Equation (38):

$$C_{k+1} = \eta_c C_k + \beta_1 \exp(-\beta_2 / \Delta t_k) \tag{38}$$

where C_k is the charging capacity in the k cycle, Δt_k is the rest time interval from the k cycle to the $k+1$, β_1 and β_2 are the parameters to be identified. Similarly, the capacity degradation trend can also be used to evaluate SOH.

Literature (An et al., 2013) proposes a hybrid methods based on an empirical degradation model-based method and a PF method, where the PF updates parameters. SOH or RUL can be predicted by constantly updating and predicting state parameters (internal resistance, capacity and so on). Among them, the equations of state and observations established are:

$$\begin{cases} x_k = f(x_{k-1}, \theta_k, v_k) \\ z_k = h(x_k, w_k) \end{cases} \tag{39}$$

where k is the time step index, x_k is the damage state, θ_k is a vector of model parameters, z_k is measurement data. v_k and w_k are, respectively, process and measurement noise. In the prognostics area, the state transition function f is referred to as a damage model, which can be expressed based on the exponential growth model as shown in Equation (40):

$$f = a \exp(-bt) \tag{40}$$

where a and b are model parameters, t is time or cycles, and λ is the internal battery performance which is stated as either the electrolyte resistance.

As to the combination of the same type SOH prediction methods, hybrid methods between data-driven methods are the most common. For example, in literature (Chen et al., 2019b), the authors propose a fusion model based on the ARMA and Elman to achieve SOH prediction. First, the EMD algorithm is applied to decompose the SOH variation signal into several intrinsic mode functions (IMFs) and residual. Then, the ARMA model and Elman neural network model are respectively built by training the IMFs data and residue data. Finally, all the individual predictions are combined to generate the estimated SOH sequences. Among them, the input of Elman is the extracted aging feature (the constant charging duration), and the output is the residual. Zheng and Fang (2015) use a hybrid method between RVM and UKF to predict the capacity in the short-term and improve prediction accuracy. Firstly, An RVR model is employed as a nonlinear time-series prediction model, then the input and output variables are constructed by reconstructing the data, its form is similar to Table 4. Finally, the predicted filter residual is then fed into the UKF correction step for future prediction. The literature (Wu et al., 2019) proposes a hybrid method for predicting the RUL combined with ANN and PF. Firstly, an ANN with better performance than the typical empirical degradation model is introduced to simulate the degradation trend under different operating conditions. The capacity degradation formulation as shown in Equation (41). In NN, the input is cycling number and the output is the battery capacity. Then the parameters of the ANN model are recursively updated with PF. Among them, the bat algorithm is used to improve the PF, which achieves higher prediction accuracy. As described before, this method can also predict SOH.

$$Q = \varphi_1 \cdot \exp(\varphi_2 \cdot C) + \varphi_3 \cdot \exp(\varphi_4 \cdot C) \tag{41}$$

where φ_1, φ_1, φ_1, φ_1 are the model parameters.

For the combination of optimization algorithm and other methods, Zhang (Zhang et al., 2016) uses PSO based radial basis function (RBF) neural network algorithm to predict SOH for the combination of optimization algorithm and other methods. Among them, the input is: constant current charging time, constant voltage charging time, instantaneous voltage sag value, constant discharge time, static 1-min voltage recovery value. For predicting the SOH in mine movable rescue capsule precisely, literature (Li et al., 2012) establishes the model to predict the SOH of lithium iron phosphate (LFP) battery by Elman neural network, optimizes its original weights and threshold by genetic algorithm (GA), and calculates SOH by shallow discharge testing data. The literature (Xiao et al., 2017) uses the internal resistance method to define the SOH, and a new ANN algorithm optimized by the ant colony algorithm is proposed. Among them, the ant colony algorithm optimizes the weights and thresholds of the ANN. Charge and discharge time, battery cycle times and DOD are used as the input of the ANN, and the internal resistance is used as the output of the model, thereby predicting SOH.

5.2.4. Other methods

We systematically introduced the model-based SOH prediction method, the data-driven SOH prediction method, and hybrid method before this section. Although we try to classify the current SOH prediction methods as much as possible clearly, there are still a lot of methods that cannot be generalized by these three major methods, so the prediction methods that do not belong to these three categories are called other methods. According to the summary and analysis, other methods are divided into based on SOC, IC method, and other novel methods in this section.

Among them, in the previous model-based, data-driven, and hybrid method-based SOH prediction methods, we can see that some SOH evaluation methods have introduced SOC, which can be a certain relationship between SOC and SOH from a certain point of view. For example: In the model-based (Literature (Paul et al., 2013)) and data-driven methods (Literature (Qin et al., 2017; Kim, 2014)), SOC is used as a feature index as an input to the SOH evaluation model. Besides, we can see from the literature (Cheng et al., 2008; Yan et al., 2019; Fang et al., 2019; Guo et al., 2017; Kim et al., 2012; Chen et al., 2014; Chen and Wang, 2014; Wang et al., 2016) that in the filtering-based methods, the terminal voltage of the battery is usually used as the observation equation, while the OCV in the observation equation can be used as SOC evaluation. So the SOC-based prediction method mentioned in this section refers to the calculation of the battery capacity for a certain period of time (or the maximum available capacity) by the ampere-hour integration method or the OCV method, which is compared with the corresponding capacity of a fresh battery to obtain SOH. This type of method is simple and is also a commonly used SOH prediction method, so we will introduce the SOC-based prediction method independently.

5.2.4.1. SOC-based method. As can be seen from the previous introduction, the SOC-based method we mentioned in this paper estimates the SOH by the measurement of the corresponding battery capacity by the ampere-hour integration method or the OCV method and then achieves real-time prediction of SOH by a series of control strategies. The core formula for this type of method by using the ampere-hour integration method is shown in Equation (42).

$$\int_{t_1}^{t_2} \frac{\eta i(t)}{3600} dt = C(SOC(t_2) - SOC(t_1)) \tag{42}$$

where the C denotes the battery capacity, η is the columbic efficiency, t_1 and t_2 respectively represent the start time of charge or discharge.

The literature (Capitaine and Wang, 2018) presents a novel design for a test platform to determine the SOH, data acquired from the testing circuitry are stored and displayed in LabVIEW to obtain the charging and discharging curves. Finally, the SOH of the battery is then calculated using a Coulomb counting method in LabVIEW. In literature (Le and Tang, 2011), the coulomb counting method and the OCV are used to calculate SOH. The essence is to use the two methods to obtain the maximum charge and discharge electric quantity, and then reflect the SOH through the maximum charge and discharge electric quantity. The experimental results show that the coulomb counting method better than the OCV method.

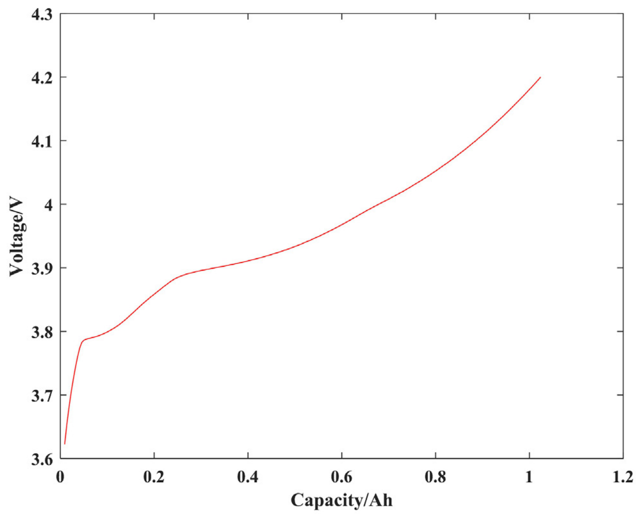
5.2.4.2. Incremental capacity method. Besides, some other SOH prediction methods can be considered separately. We call this method the correlation characteristic prediction method of charge-discharge experimental curve, the most classical method of SOH prediction by extracting the characteristics of the IC curve and DV curve (In a way, PF also belongs to), then the characteristics are combined them with other algorithms.

The IC is replaced by a very clever process that uses the form of curve which dQ/dV ~ V instead of the traditional curve of V ~ Q. The traditional V ~ Q curve changes only significantly at the beginning and the end of the voltage and changes slowly in the middle of the voltage, but the middle of the voltage is exactly the period where the chemical reaction of the battery occurs. For charging and discharging of the battery, the medium-term charge of voltage and chemical reaction are the most, so the study of V ~ Q curve for battery is unreasonable, but the IC curve amplifies ingeniously the medium-term response of voltage through the differentiation of voltage and capacitance.

The comparison of the IC curve with the V ~ Q curve is shown in Fig. 11. Based on the above analysis, we can see that the IC curve can extract more obvious features, and their characteristic is closely related to the internal reaction and mechanism of battery aging for the LIBs, so it can be used for further research. Li et al. (2019c) use the incremental capacity analysis (ICA) method to extract the curve, then filter the IC curve to better extract important health characteristic indexes. Finally, they use the GPR method to predict SOH. Among them, all features to be selected every 30mv from 3.8V to 4.1V, and the 6 features with the highest correlation are selected as the input of the GPR model through correlation analysis. In literature (Li et al., 2019d), Grey relational analysis (GRA) and the entropy weight method (EWM) are then used to establish a battery degradation model based on the health performance indicators. Among them, for battery SOH estimation based on the IC curves, the IC curve derived from new cells is defined as the reference for SOH evaluation.

5.2.4.3. Novel method. In addition to the above-mentioned SOH prediction methods, there are some relatively novel and ingenious methods. These methods are analyzed from other angles to predict SOH and obtain good prediction results. In literature (Kim, 2010), the authors use double sliding the mode observer to estimate SOC. Among them, slow time-varying observer estimates SOH according to the decay of battery capacity and the change of internal resistance, while fast time-varying observer estimates SOC, terminal voltage and polarization effect inside the battery.

B. Sun combines the Delphi method with the grey correlation analysis for SOH estimation in literature (Sun et al., 2015). He considers six main factors affecting SOH. Among them, the experts determine the weight of the six factors by scoring them, and then he uses the GRA to optimize the parameters affecting these factors. Among them, the grey relational grade analysis (GRGA) model is expressed as in Equation (43):



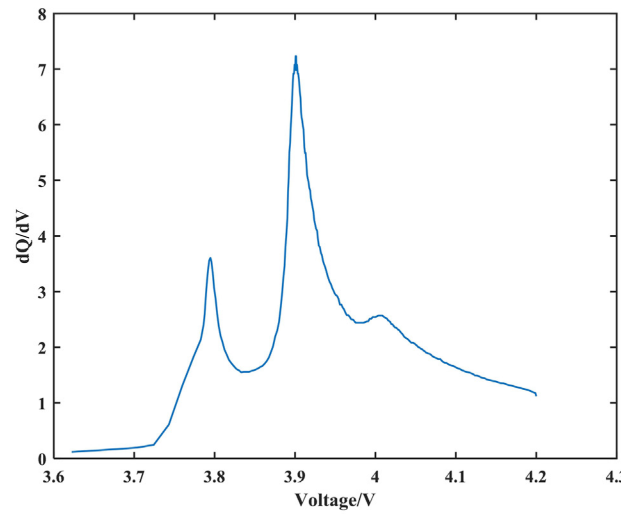


Fig. 11. (a) The curve of V ~ Q. (b) The curve of dQ/dV ~ V.

$$R = E \times W^T \tag{43}$$

where R is an assessment vector representing the synthesized results. W the weighting vector, and E the judgment matrix.

In literature (Kim and Cho, 2014), the authors present a new approach for characteristic analysis based on the discrete wavelet transform (DWT) to predict SOH. Firstly, they decompose the voltage signal into n decomposition level (approximation components and detailed components), the optimum number of resolution levels in the analysis is chosen. Then, they choose characteristics from the best approximation components and the best detailed components. Finally, the SOH evaluation equation is:

$$\underset{arbitrary}{SOH} = \frac{\left|\frac{A_5^{curr} - A_5^{aged}}{A_5^{fresh} - A_5^{aged}}\right| + \left|\frac{D_5^{curr} - D_5^{aged}}{D_5^{fresh} - D_5^{aged}}\right|}{2} = \frac{SOH_{A_5} + SOH_{D_5}}{2} \tag{44}$$

where A_5^{curr} and D_5^{curr} are the standard deviation of approximation A_5 detail D_5 components for an arbitrary LIBs respectively, $A_5^{fresh}(D_5^{fresh})$ and $A_5^{aged}(D_5^{aged})$ are each standard deviations approximation A_5 detail D_5 components for fully fresh and aged LIBs in 1.3 Ah cell group.

Chen et al. (Chen et al., 2019c) aim at the problems of a complex algorithm, complicated programming and difficult development of single-chip in the SOH prediction process. A SOH prediction method is proposed, which uses a single chip as the main chip and the number of LIBs charging and discharging cycles as the benchmark index of SOH. The influence on SOH is converted into the number of cycles, the non-linear relationship between the main factors and SOH is made into a two-dimensional array table. In literature (Winodo et al., 2011), the authors use the sample entropy (SampEn) of the battery discharge voltage as the input data and directly implement the SOH prediction by using the RVM algorithm.

So far, we have made the current research status of SOH prediction as clear as possible. According to the previous introduction, the SOH prediction method summarized in this paper is as shown in the following Fig. 12, and a detailed synopsis of the evaluation results for the SoH/RUL estimation methods is provided in Tables 5–7.

According to the previous introduction, we propose a classification framework for SOH prediction methods, so that the current research on SOH can be presented to readers in a clear and orderly manner.

6. Evaluation of SOH prediction methods

There are currently many SOH prediction methods. Many researchers claim their methods are better. Tables 5–7 show the prediction accuracies in many of these methods are all quite high, which can bring confusion to the discussion. However, all the proposed SOH prediction methods have their scope of application, and better results can be achieved under certain circumstances. Also, the complexity and the mode of operation for each method are different, which can affect actual applications. Therefore, it is necessary to evaluate the SOH prediction methods, as discussed in Section 5, to understand the scope of application and the complexity of these methods and serve as a reference for actual implementation and for future research. In this section, we analyze the benefits and drawbacks of existing SOH prediction methods, provide commercial applications combined with the proposed SOH prediction methods and their descriptions (Tables 5–7), and give the evaluation results, as shown in Fig. 13.

Based on earlier discussions, most of the methods have not been widely used in industry and commerce. The general SOH estimation technology is weak, which is why the research on battery SOH is less than that of SOC. In the next section, we will combine the actual operating conditions of BEVs, highlight the problems and challenges in some of the methods, and propose corresponding suggestions and solutions.

7. Challenges and recommendations for SOH prediction methods

As described in the SOH prediction status in Section 5 and the evaluation of the SOH prediction methods in Section 6, we can find that there are numerous SOH prediction methods. The major direction for improvements in SOH prediction is geared towards the hybrid method, with the primary goal of obtaining higher accuracy. Likewise, the environment and the operating conditions can easily influence the results of SOH prediction. In this section, the current

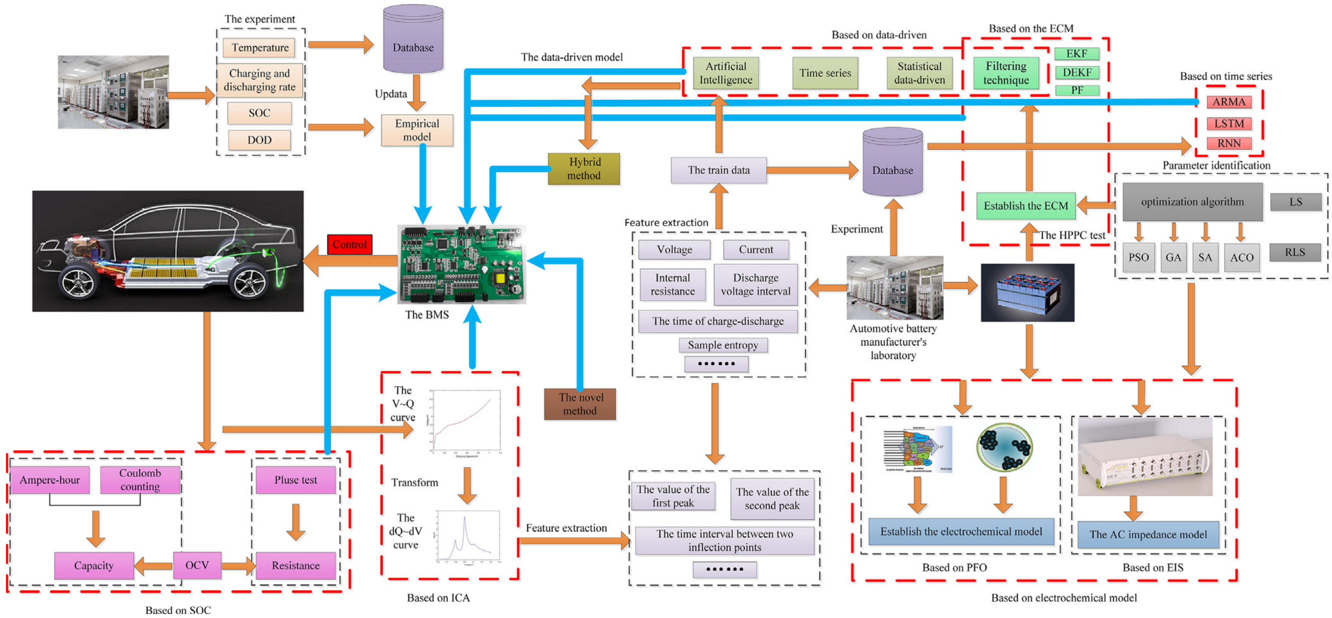


Fig. 12. The methods of SOH prediction.

Table 5
A detailed synopsis of the evaluation results for the SOH/RUL estimation methods based on model.

Method	Refs	Battery chemistry	SoH/RUL estimation	Data of Calculation	Mode of operation	Evaluation index	Estimation precision
ECM-based method	Zhou et al. (2019)	Battery pack	SOH	Matrix operation, Function and exponential expressions	Online	Accuracy	<8%
Aging mechanism-based method	Prasad and Rahn (2013)	LIBs	SOH	Differential equation, transfer function and Matrix operation	Offline	Best fit values	99%
	Safari and Delacourt (2011)	LFP	SOH	Matrix operation, Function and exponential expressions	Offline	The results	In line
	Li et al. (2018a)	$LiFePO_4$/Graphite battery	SOH	Matrix operation	Offline	RMSE	1.03×10^{-4}
EIS-based method	Xia and Abu Qahouq (2019)	Tenergy 30005-0	SOH	Function expressions and model	Online	N/A	N/A
Empirical degradation model-based method	Singh et al. (2019)	NMC	SOH	Exponential expressions	Offline	The maximum estimated difference	1.2%
	Paul et al. (2013)	$LiFePO_4$/Graphite	SOH	Exponential expressions	Online	maximum error	About 8%
	John et al. (2011)	$LiFePO_4$/Graphite	SOH	Exponential expressions and other expressions	Offline	N/A	The model projections are consistent with the experimental data
	Su et al. (2016)	$LiNi_xCo_yAlxO_2$/Graphite	SOH	Matrix operation and other expressions	N/A	Accuracy	92%
Probabilistic model-based method	Feng et al. (2013)	$LiMn_2O_4$	SOH	Statistics	Online	Error	5%

status of the SOH prediction methods is discussed, and recommendations are proposed by combining the actual BEVs.

7.1. The method based on the ECM

The current ECM, selected by most literature, is the traditional Thevenin or the second-order RC model. These methods are modeled and predicted under the conditions of simulation experiments. Among them, the battery is described by any number of RC elements without considering the electrochemical factors, which allows for speedy calculations and simple implementations (Krewer et al., 2018). The current status and challenges in the actual operating environment are shown in Fig. 14.

According to the above description, the standard ECM parameters lack physical and chemical meanings, and the prediction effect is poor in the actual BEVs operating environment. Thus, we can consider an ECM based on electrochemical processes. ECMs that are derived from electrochemical effects and processes are usually used to optimize the design of battery systems, which considers energy content, cooling, and performance, as well as for concept evaluations (Krewer et al., 2018). Its basic composition and schematics are shown in Fig. 15.

As shown in Fig. 15, the starting point in modeling is a sectional view of the cell, and the electrical elements are drawn representing the physical effects and processes. In addition, the model structure is created using the recorded cell impedance (see Fig. 15), and the

Table 6
A detailed synopsis of the evaluation results for the SOH/RUL estimation methods based on data driven.

Method	Refs	Battery chemistry	SoH/RUL estimation	Data of Calculation	Mode of operation	Evaluation index	Estimation precision
ANN	Yang et al. (2016)	$LiFePO_4$/Graphite	SOH	Matrix operation and model	Online	The maximum error	<8%
	Pan et al. (2018)	IFP1865140 type LIBs	RUL	Matrix operation and model (FFNN)	Online	Error	<5%
	Rezvain et al. (2011)	LIBs	SOH	Matrix operation and model (AdNN)	Offline	MSE	$<2 \times 10^{-4}$
SVM	Nuhic et al. (2013)	High power LIBs	SOH and RUL	Matrix operation and model (SVM)	Online	The performance	Good
	Pattipati et al. (2011)	Second-generation LIBs	SOH	Matrix operation and model (SVM)	Online	Error	About 2%
	Chen et al. (2018a)	LiFePO4/Graphite	SOH	Matrix operation and model (LS-SVM)	Offline	RMSE	0.32%
RVM	Qin et al. (2017)	LIBs	RUL	Matrix operation and model (RVM)	N/A	RMSE	<5%
	Li et al. (2014b)	LIBs	SOH	Matrix operation and model (RVM)	Offline	RMSE	<0.004
Fuzzy logic method	Chen et al. (2019a)	18650 ternary LIBs	SOH	Functional operation and model (T-S)	Online	Maximum prediction Error	4.3%
	Kim (2014)	$LiMn_2O_4$/Graphite	SOH	Functional operation and model (fuzzy logic-controlled)	Online	N/A	clearly appropriate
Integrated learning	Li et al. (2019b)	LIBs	SOH	Matrix operation and model (AdaBoost.RT)	N/A	RMSE	<3%
	Chen et al. (2018b)	LIBs	SOH	Matrix operation and model (SVR and AdaBoost.RT)	N/A	MAPE	<2%
EKF	Cheng et al. (2008)	$LiFePO_4$/Graphite	SOH	Matrix operation and equation set	Online	N/A	Accurate estimation
	Yan et al. (2019)	Sony 18650 LIBs	SOH	Matrix operation and equation set (LS_EKF)	Online	N/A	The LS_EKF is better than the traditional RS-EKF
DEKF	Fang et al. (2019)	LIBs	SOH	Matrix operation and equation set (DEKF)	Online	The maximum estimation error	1.52%
	Guo et al. (2017)	Lithium iron phosphate battery	SOH	Matrix operation and equation set (DEKF)	Online	The maximum error	<4%
UKF	Kim et al. (2012)	LIBs	SOH	Matrix operation and equation set (DEKF)	Online	Error	<5%
	Chen et al. (2014)	US18650V3 LIB	SOH	Matrix operation and equation set (UKF)	Online	Error	<4%
	Chen and Wang (2014)	18650 LIBs	SOH	Matrix operation and equation set (UKF)	Online	Estimation error	<5%
	Wang et al. (2016)	18650 LIBs	SOH	Matrix operation and equation set (UKF)	Online	N/A	N/A
PF	Dong et al. (2018)	LIBs	SOH	Matrix operation and equation set (PF)	Online	RMSE	<4%
	Dong and Jin (2014)	18650 LIBs	SOH	Matrix operation, exponential expressions and equation set (PF)	Online	Error	<7%
	Zhang et al. (2014a)	TBP0306 LIBs	SOH	Matrix operation, exponential expressions and equation set (PF)	N/A	Residual error	<5%
GPR	Peng et al. (2018)	LIBs	RUL	Matrix operation, exponential expressions and model (HGFPR)	Online	Accuracy	2.2%
	Liu et al. (2013)	18650 LIBs	SOH	Matrix operation, exponential expressions and model (QGFPR)	N/A	MAPE	<0.1%
WP	Xu et al. (2019)	LIBs	SOH	Matrix operation, exponential expressions and model (WP)	Online	MSE	$<3 \times 10^{-3}$
ARMA	Long et al. (2013)	LIBs	RUL	Matrix operation, and model (AR)	Online	Error	12.3%
	Zhou and Huang (2016)	LIBs	RUL	Matrix operation, and model (EMD and ARIMA)	Online	RMSE	<.0.05
RNN	Eddahech et al. (2012)	$LiNi_xCo_yAl_zO_2$/graphite	SOH	Matrix operation, and model (RNN)	Online	MSE	<5%
LSTM	Qu et al. (2019b)	LIBs	SOH	Matrix operation, and model (RNN)	Online	RMSE	<0.02%
	Chen et al. (2018c)	LIBs	RUL	Matrix operation and Probability calculation	N/A	RMSE	<4%

model order is reduced by combining the elements. One of the main advantages of the presented approach is its modular design, which allows an extension to further cell effects, such as hysteresis, reversible heat generation, and temperature dependence of the OCV (entropy change), as well as diffusion-limiting effects.

The elements of the ECM and their correlation to physical processes, as shown in Fig. 15, can easily be combined. However, the transformation of these elements into the time domain is not trivial. Previous studies have explored possible approximations and transformation of some of these elements into the time domain (Jossen, 2006). At the same time, it can easily be extended to a battery system model. In Fig. 16, a standard model approach for a battery system is presented.

The battery model consists of the electrical ECM, the thermal

Table 7
A detailed synopsis of the evaluation results for the SOH/RUL estimation methods based on Hybrid and other methods.

Method	Refs	Battery chemistry	SoH/RUL estimation	Data of Calculation	Mode of operation	Evaluation index	Estimation precision
Hybrid methods	Zenati et al. (2010)	LIBs	SOH	Matrix operation, and model (GM and PF)	Offline	N/A	N/A
	Zhao et al. (2019)	LIBs	RUL	Matrix operation, and model (GM and SVR)	Online	RMSE	<5%
	Liu et al. (2014)	LIBs	RUL	Matrix operation, and model (PF and AR)	Online	RMSE	<2%
	Chen et al. (2019b)	Li $(NiCoMn)O_2$	SOH	Matrix operation, and model (EMD and AR)	N/A	MSE	$<1 \times 10^{-4}$
	Zheng and Fang (2015)	LIBs	SOH	Matrix operation, and model (RVM and UKF)	Online	RMSE	<3%
	Wu et al. (2019)	LIBs	RUL	Matrix operation, exponential expressions and model (NN and PF)	N/A	Error	About 10%
	Zhang et al. (2016)	18650 LIBs	SOH	Matrix operation and model (RBF)	Offline	RMSE	2.81%
	Li et al. (2012)	LFP	SOH	Matrix operation and model (Elman)	Offline	Standard error	<5%
	Xiao et al. (2017)	LFP	SOH	Matrix operation and model (ANN)	Offline	Average error	0.7%
Based on SOC	Capitaine and Wang (2018)	18650 battery	SOH	integral operation	Online	Average deviation	0.56%
	Le and Tang (2011)	$LiFePo_4$/Graphite	SOH	Matrix operation and differential operation	Online	The maximum estimation error	<4%
Incremental capacity method	Li et al. (2019c)	LIBs	SOH	Matrix operation and model (GPR)	Online	RMSE	<0.001
	Li et al. (2019d)	$LiFePO_4$/Graphite	SOH	Matrix operation and model (GRA and EWM)	Online	The maximum error	<4%
Novel method	Kim (2010)	$LiMn_2O_4$	SOH	N/A	Online	Error	<3%
	Sun et al. (2015)	$LiMn_2O_4$	SOH	Matrix operation and model (GRGA and DM)	Online	N/A	N/A
	Kim and Cho (2014)	Samsung SDI 18650 $LiCoO_2$ LIBs	SOH	Matrix operation and model (DWT)	Online	N/A	SOH evaluation can be implemented
	Chen et al. (2019c)	LIBs	SOH	N/A	Online	MRE	4.6%
	Winodo et al. (2011)	LIBs	SOH	Matrix operation and model (RVM and SampEn)	N/A	RMSE	$<1 \times 10^{-4}$

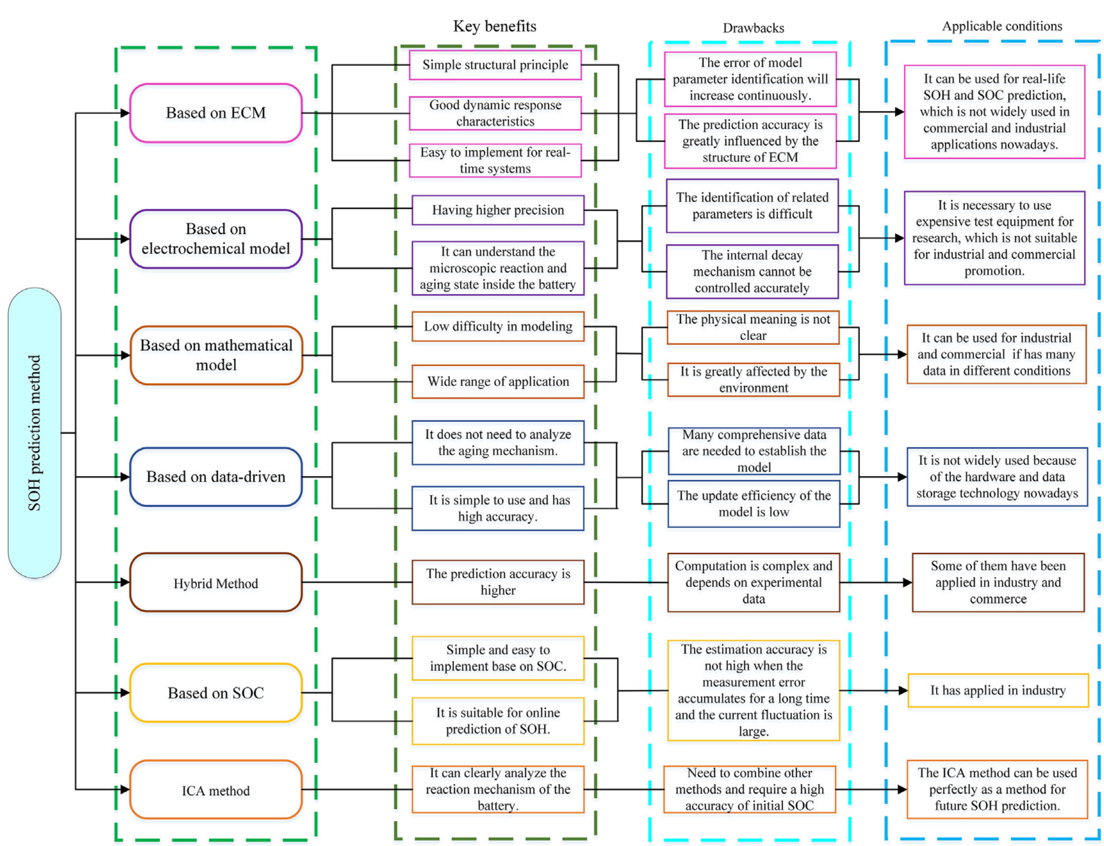


Fig. 13. The evaluation results of SOH prediction methods.

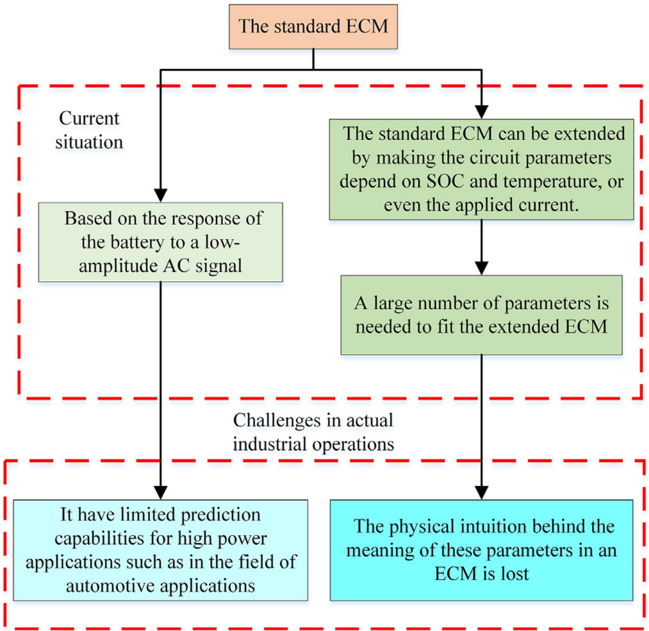


Fig. 14. The actual operating environment and challenge of standard ECM.

model, and the model parameter sets. Such models allow online monitoring and control of batteries. Due to the interaction of thermal and electrical processes in the model, battery behavior can be predicted under a wide range of operating conditions (Krewer et al., 2018).

7.2. The method based on the data-driven

The data-driven method is explained in two parts.

The first aspect is the prediction method based on feature input, such as ANN, SVM, RVM, and ensemble learning. They can also be used as time series-based prediction methods, but a reconstruction of time series-based data is required. Fig. 17 shows the basic workflow required for online SOH estimation based on feature input prediction methods. Among them, feature extraction is a crucial step and challenge in SOH estimation (Li et al., 2019e), which directly affects performance, and is therefore added to the workflow.

According to the previous description, obtaining many external features during operation is difficult due to the limited computing power of existing BMS. Therefore, the SOH monitoring method is highly desirable, given that it can directly use the direct external features of the battery for SOH estimation. In addition, it is advisable to avoid battery modeling and data preprocessing steps to reduce the calculation workload (Li et al., 2019e).

The second part is based on time series prediction methods, such as GPR, WR, AR, RNN, and LSTM. Among them, AR, RNN, and LSTM are non-probabilistic methods, Non-probabilistic data-driven approaches can only provide an estimated point in regression. However, capturing the prediction uncertainty level of the conditional distribution is a real challenge due to uncertainties from various sources, such as measurements, state estimation, model inaccuracies, and future load uncertainty (Goebel et al., 2008). In addition, the battery ages faster, and its prediction accuracy becomes lower as the prediction time increases. An adaptive correction trend factor can be added in order to make the predictions more accurate. On the other hand, the GPR and WR are probabilistic methods with the ability to yield PDFs, predict data points, and provide confidence bounds around them. Because of these, probabilistic data-driven methods are preferable, given that the uncertainty estimation can benefit battery users. However, the development of probabilistic data-driven methods is still in its infancy. These models are obtained under the same training conditions, which raises questions about their robustness in practical applications, as actual operating conditions may vary widely. It is therefore recommended to improve probabilistic techniques by training the models under complex aging conditions. Additionally, the performance of these techniques is also highly sensitive to their structure and parameters. Suitable structure determination and parameter optimization strategies should also be explored to enhance their performance for future self-adaptive health or lifetime prediction (Li et al., 2019e).

According to the previous introduction, good modeling is vital to forecasting. Similarly, updating the model is also very important because the model built offline would not be able to include all kinds of complex working conditions. Therefore, the model needs to have update functions to meet the dynamic running environment. In the actual BEVs system, the BMS accepts a large amount of high-speed data, which has high requirements on data storage and

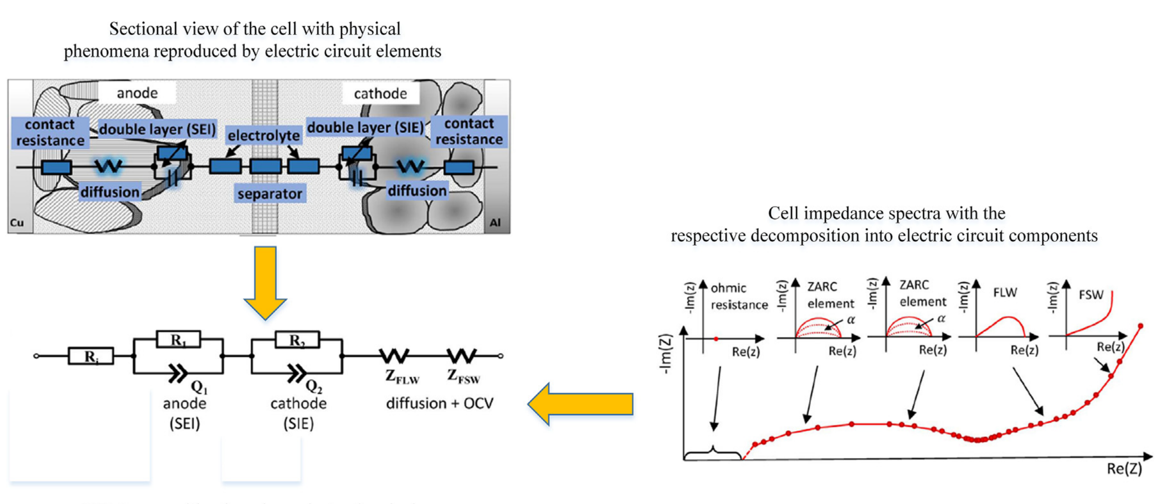


Fig. 15. A typical procedure for creating an electrochemical ECM (Literature (Krewer et al., 2018)).

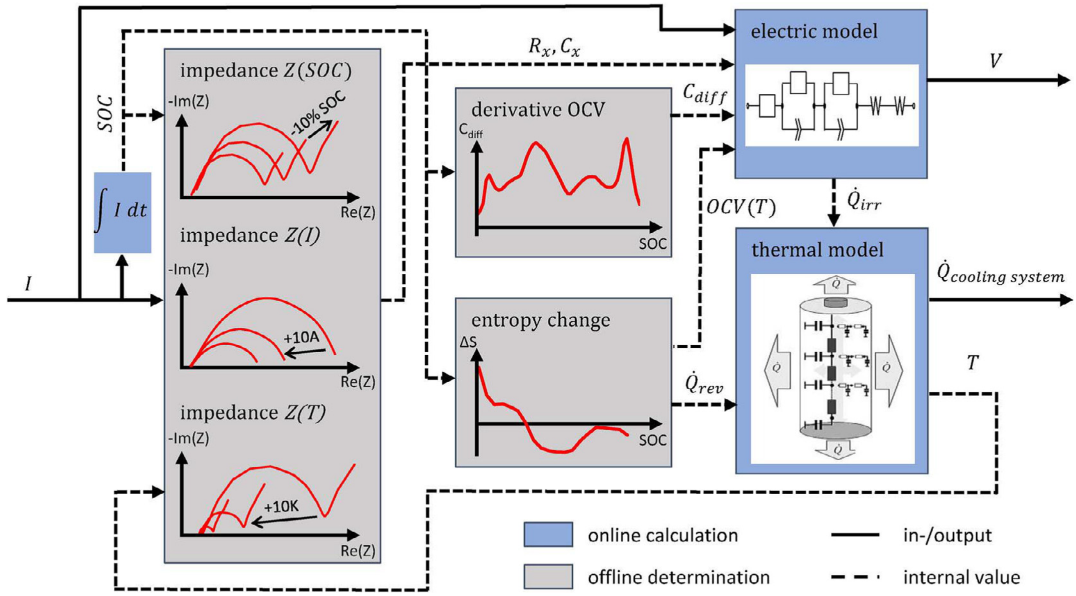


Fig. 16. The common model approach for a battery system (Literature (Krewer et al., 2018)).

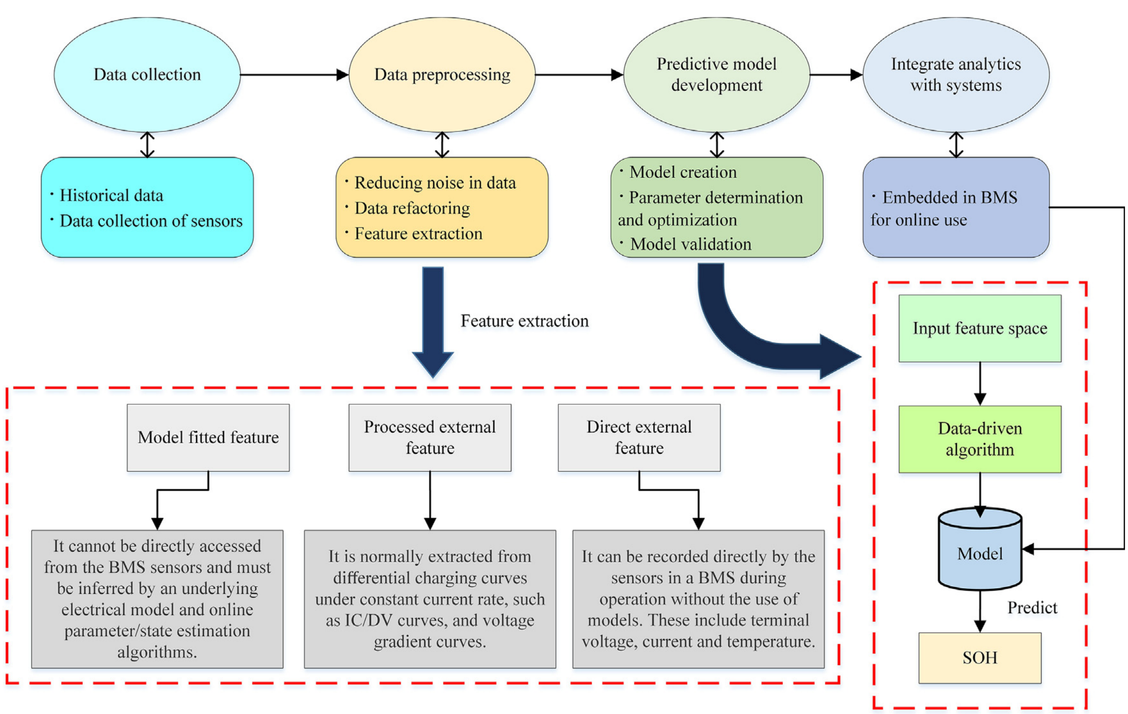


Fig. 17. The basic workflow required for online SOH estimation based on feature input prediction methods.

internal computing of the BMS. The model embedded in the BMS cannot always use these data in calculations or updating. Thus, a strategy to update the model becomes highly necessary (Tian et al.,). The schematic diagram of the proposed model update strategy is shown in Fig. 18.

If a well-established initial model is embedded into the BMS, the concept drift can be used to guide updates for the model, such that if the concept drift occurs at the given moment, the model will be updated. Otherwise, the model will not be updated, which will significantly reduce the required time for model updates.

7.3. The method based on ICA

IC/DV analysis provides a non-destructive means of characterization of cells and has been widely used for aging mechanism identification (Dubarry et al., 2011). Its hardware requirement is low, and the characteristics extracted from the IC/DV curve can reflect the internal reaction mechanism of the battery very well. However, it still has specific problems in practical applications. The challenges with ICA/DVA are presented in Fig. 19.

The ICA/DVA is a powerful apparatus for online SOH estimation (Wang et al., 2017) and can be easily implemented in BMS by

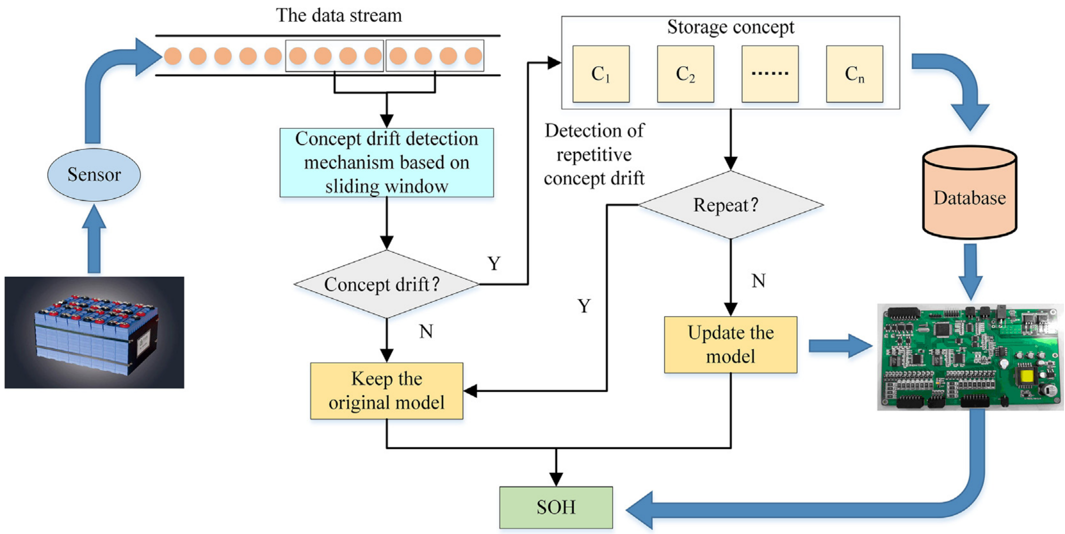


Fig. 18. The schematic diagram of the model update strategy.

monitoring only two parameters (voltage and charge and discharge capacity). However, it still faces three main challenges in actual production. In order to overcome the first challenge, we can convert the IC/DV curve into a PDF curve (Feng et al., 2013), which can be solved by calculating the probability of occurrence of the different voltage value points in the battery charge and discharge curve. To solve the second problem, the SOH estimation method based on ICA/DVA should be able to use part of the charging voltage curve (Li et al., 2018b). The basic approach is shown in Fig. 19. In response to the third problem, we can add some circuit elements in the actual circuit, such as transformers and relays, and then use them in converting into a larger charging current, which is suitable for ICA/DVA.

The ICA/DVA method is used to identify features from the differential curve of electrical parameters during battery cycling. The concept of ICA can be combined with the temperature measurement to identify features from the differential curve of thermal or mechanical parameters. This method is called differential thermal voltammetry (DTV), which allows us to avoid the problem of the differential curve of the above electrical parameters. DTV technology detects the temperature of the battery surface during constant current charging. DTV is obtained by differentiating temperature (T) from the voltage (DT/DV) and then plotting the battery voltage. DTV is designed to easily and quickly reveal the most obvious entropy-related information during cell manipulation (Li et al., 2019e). The DTV expression is presented in Fig. 20.

Generally, DTV is considered as experimentally easy and is applicable for parallel-connected cells with the advantage of enabling higher current rate tests than the ones required for IC/DV analysis. It only requires monitoring the parameters of voltage and temperature during the galvanostatic charge/discharge process, which shows great potential for online applications (Li et al., 2019e). Moreover, DTV does not require strict isothermal conditions, meaning that there is no need for super-efficient cooling of batteries, making analysis experimentally easier. Based on the above description, the ICA solution is shown in Fig. 21.

7.4. The method based on the SOC

Many approaches for SOH estimation use the method based on the SOC. The ideas and principles behind SOH estimation are simple and straightforward. The estimates are calculates based on the relationship between Ampere-hours (charged or discharged from the battery) and the difference between the two values of the

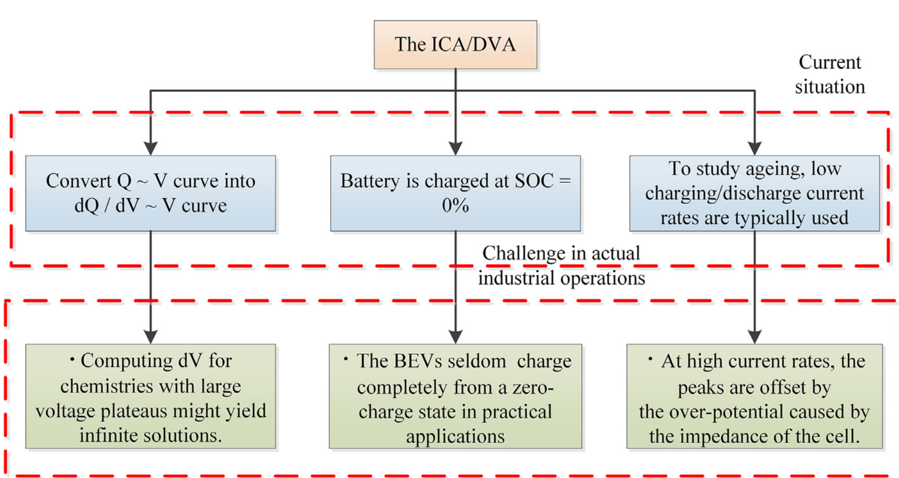


Fig. 19. The challenge with ICA/DVA.

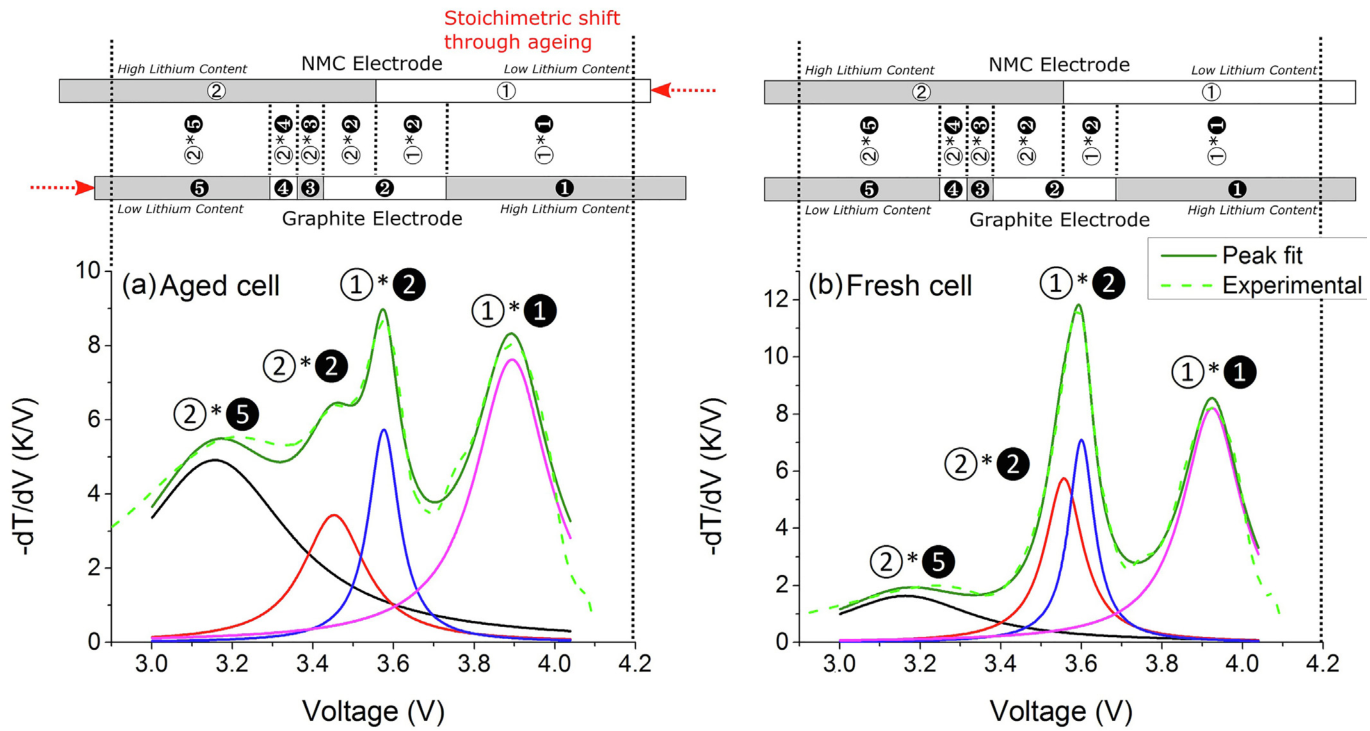


Fig. 20. The DTV expression (Literature (Merla et al., 2016)).

voltage-based SOC (or voltages) relative to the initial and final points where Ampere-hour throughput is measured (Farmann et al., 2015b). Fig. 22 shows the basic concepts and principles for the SOC-based SOH estimation.

Although the SOH prediction method based on the SOC is simple in principle and can easily be implemented online in a BMS system, there are several challenges associated with the approach. In general, after charging or discharging the battery, the relaxation time necessary to obtain a cell in an entirely steady-state resides in the range of hours (Zhang et al., 2014a). If the SOC is determined by measuring the OCV at the current time, it will take a long time to determine the accurate SOC value, which can affect the real-time evaluation of BEVs. In order to improve and overcome these challenges, the following recommendations are proposed with the BEVs actual situation based on the SOC and OCV methods.

1. Reduce the relaxation time of the battery by analyzing the factors that influence the relaxation time. For example, less relaxation time is needed at higher temperatures than at lower temperatures because diffusion processes and chemical reactions taking place inside the cell are faster at higher temperatures than at lower temperatures, as first postulated by Fick's law (Simopoulos and Gregory, 2010).
2. Eliminate the effect of polarization reaction on the consistency of open-circuit voltage and cell voltage. According to the electrochemical principle, a single cell will generate a polarization phenomenon after charging and discharging, causing its open-circuit voltage to deviate from the battery's equilibrium voltage, which can seriously affect battery consistency. The bidirectional power amplifier is used to attenuate the sine wave. The battery is continuously charged and discharged in the positive and negative directions. After a short period of time, the sine wave attenuation is close to zero, the battery voltage is close to the target voltage, and the battery polarization phenomenon is eliminated.
3. Improve the traditional SOC and OCV joint estimation SOH method. Consider the effect of temperature, current rate, and aging factor on SOC, and improve the Ampere-Hour Method integration method.
4. Use other SOC prediction methods. For example, the effective method proposed in the literature can be used (Zhao and de Callafon, 2016; Qiu Leiet al., 2012; Liang and Wu, 2014).
5. Change the strategy based on the original method according to the general operating conditions of the BEVs to reduce the impact of relaxation time. Based on the actual BEVs operation and the charging and discharging conditions, the SOC value of the battery is corrected at the end of the battery charging, so that the accurate initial value can be obtained after each charge is completed. At the same time, the battery capacity calibration mode can be selected to calibrate at the end of the battery discharge, thereby eliminating the calculation error caused by battery aging.
6. Estimate the battery Electromotive Force (EMF) by considering OCV relaxation over only a limited period of time. A distinction is drawn between the battery OCV and EMF: any battery voltage measured under open-circuit condition is called OCV, and the equilibrium battery voltage at the end of OCV relaxation ("pure" OCV) is called EMF (Waag et al., 2014). The OCV relaxation of LIBs can be determined by the formula in Table 8.

Based on the above description, combined with the actual operation of BEVs, recommended strategies and improvements for the SOC-based SOH prediction methods are proposed and are shown in Fig. 23.

7.5. The method based on the electrochemical model

The method based on the electrochemical model uses mathematical equations based on physical and chemical knowledge to describe the processes occurring in or between the components

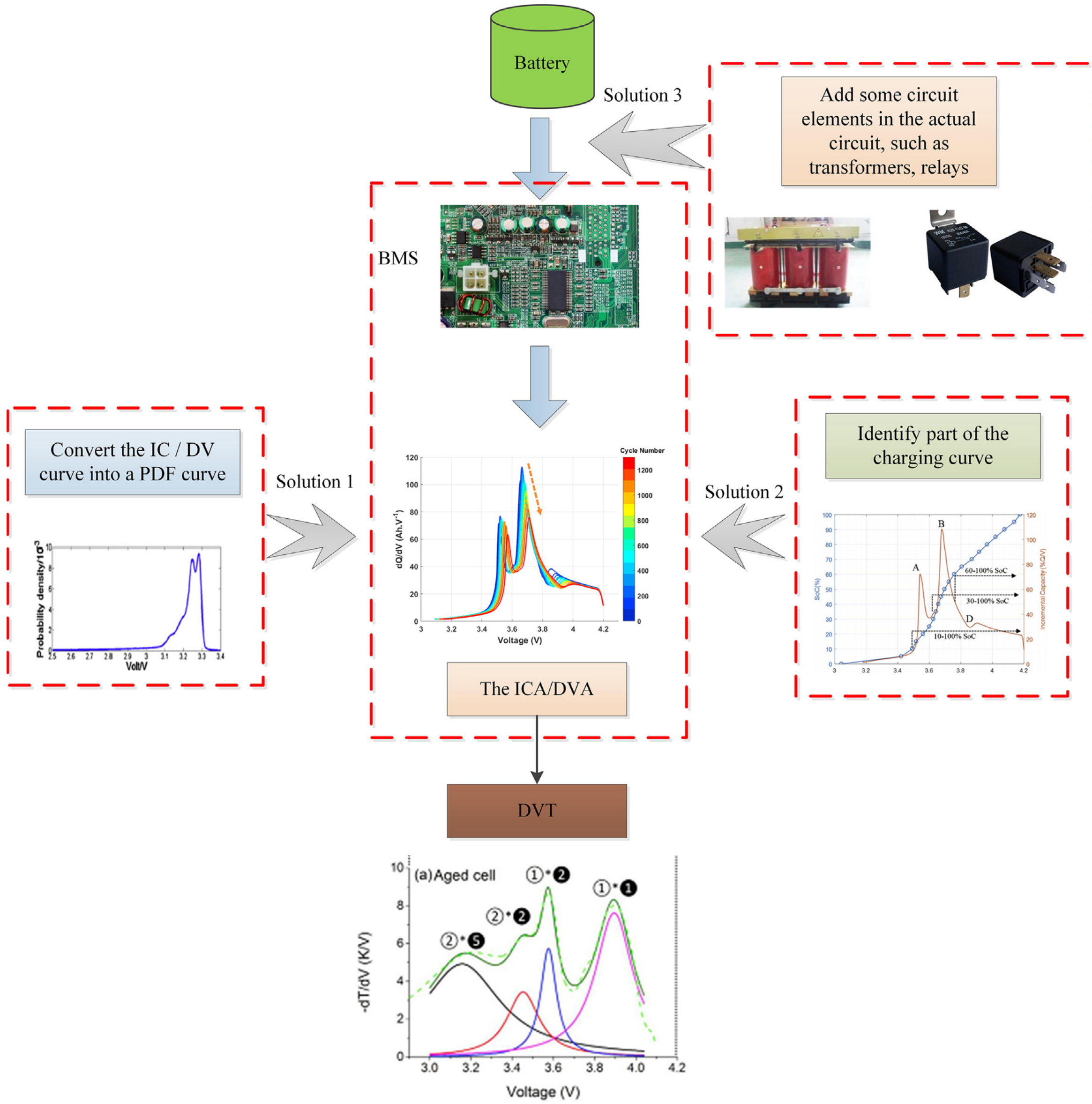


Fig. 21. The solution based on ICA.

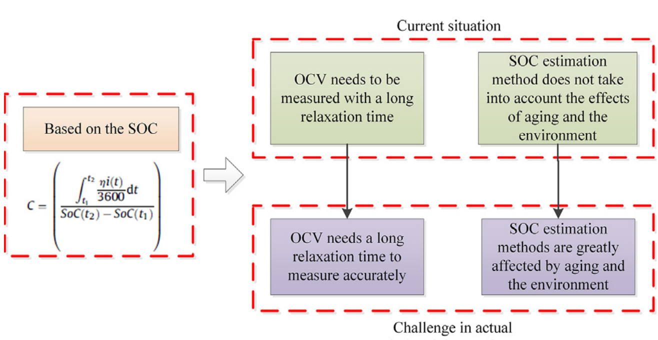


Fig. 22. The basic principle and challenge based on SOC.

and the resulting battery performance and state (e.g., SOH or SOC). Decades of research have been aimed at developing new models or model modifications that would allow more efficient computations of phenomena, more detailed physical insights, and better resolution of heterogeneity or simulation of actual geometries (Krewer et al., 2018). However, there are still numerous problems and challenges in the electrochemical model-based method. The detailed description is presented in Fig. 24.

Based on the discussion, the following observations can be made:

1. The electrochemical model or physical model of the LIBs currently used is a P2D model or a thermally coupled P2D model. These models are simulated from internal processes or external characteristics and analyzed for internal thermal behavior. However, the complete internal and external characteristic simulation of real batteries would still need to consider temperature distribution for the entire battery. Therefore, extending the description of the battery temperature distribution on the existing thermally coupled P2D model is recommended to obtain a multiphysics coupling model that would fully correspond to the physical and chemical phenomena in the

Table 8
The formula for the OCV relaxation of LIBs.

Literature	The formula
Literature (Hoenig et al., 2002)	$EMF = a \cdot V_{OCV} + b \cdot (dV_{OCV}/dt) - c \cdot T - d$
Literature (Pop et al., 2008)	$V_{OCV}(t) = EMF - \dfrac{\gamma}{t^{\alpha} \cdot \log^{\beta}(t)}$
Literature (Yang et al., 2008)	$V_{OCV}(t) = EMF - (EMF - V_{OCV}(t=0)) \cdot e^{-t/\tau}$
Literature (Waag and Sauer, 2013)	An OCV relaxation model based on theoretical consideration of the battery diffusion is proposed

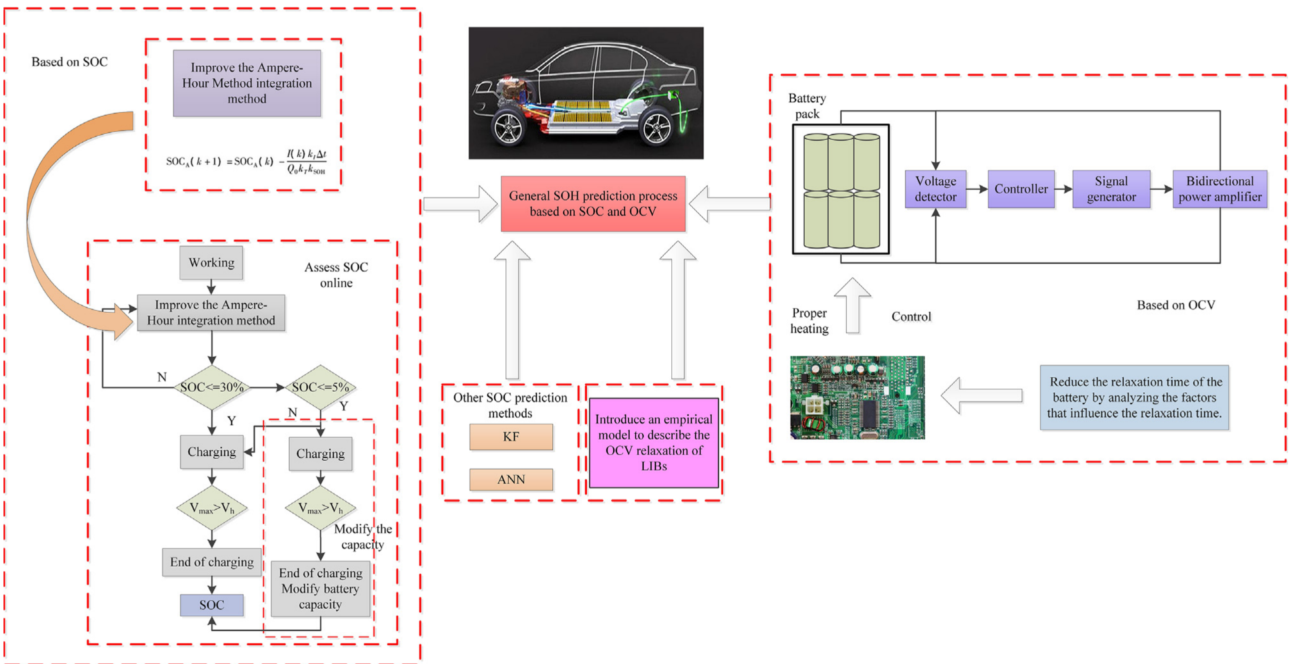


Fig. 23. The improvement and recommended strategies SOH prediction methods.

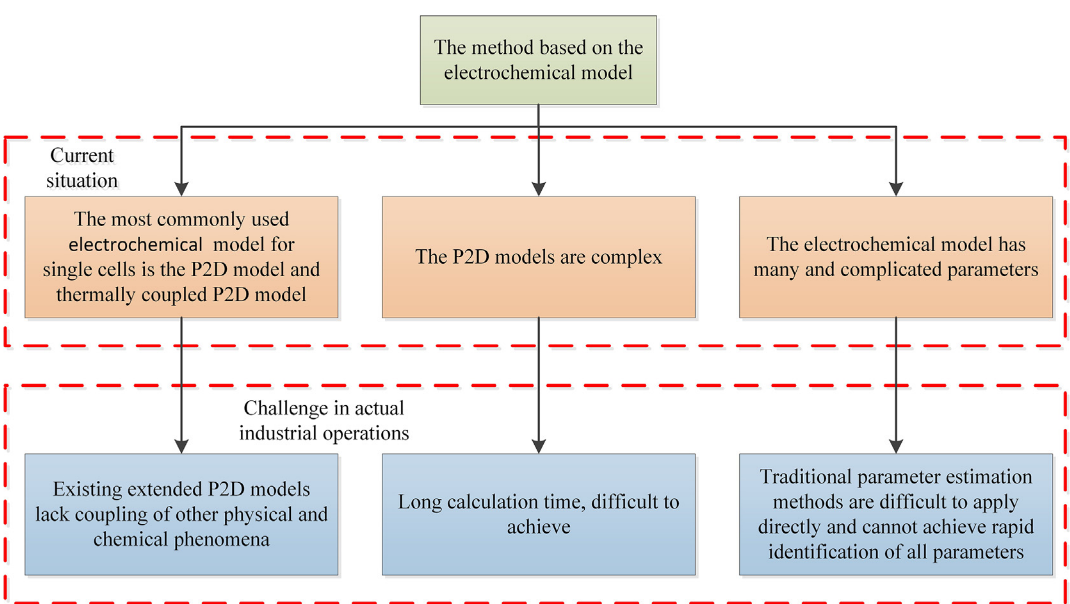


Fig. 24. The basic principle and challenge based on the electrochemical model.

real battery, from the electrode material active particle scale to the overall battery scale. This would establish the relationship between the external performance of the battery and the internal parameters of the model. Otherwise, due to volume changes in active material during lithiation/delithiation, the active material particles and electrode structures could be stressed mechanically, which can lead to cracks in particle and electrodes and loss of active material. Also, volume expansion can cause SEI destruction and change. In order to better understand the degradation in Li-ion batteries, it is recommended to focus on the interaction of various degradation processes and the impact of local conditions (Zhang et al., 2014b). The solutions and strategies for challenge 1 are shown in Fig. 24.

2. In order to make the established electrochemical model more consistent with the actual physical and chemical reactions inside the battery, extending P2D models is often more complicated, which results in higher calculation costs. Before implementing the model predictive control or state estimation, it usually becomes necessary to first simplify, such as using the single-particle model, approximating the internal diffusion process of the active substance, and ignoring the concentration gradient (Krewer et al., 2018). In addition, important parameters can be obtained through the parameter sensitivity matrix to reduce the complexities of determining multiple parameters. To decrease computational costs, another option is to reformulate the model implementation and numerics (Northrop et al., 2014; Suthar et al., 2014). This approach may facilitate their application for operation in real-time optimization in nonlinear model predictive control.
3. At present, all parameter identification methods only use battery error and measured terminal voltage error data under certain operating conditions as identification targets and do not adequately consider the effects of other influencing factors (e.g., temperature) (Zhang et al., 2013). In addition, some parameters cannot be obtained directly and can only be acquired from the manufacturer (Krewer et al., 2018). Parameters may also be sensitive to changes in the manufacturing procedure. Therefore, determining the parameters in the electrochemical model according to different working conditions and environments, and executing uncertainty quantification and global sensitivity analysis of parameters have become critical issues. Fig. 25 presents recommended strategies and improvements for determining the parameters in SOH prediction.

7.6. *The method based on the hybrid method*

The previous sections have described the challenges in and suggestions for the most common methods in SOH prediction. We have discovered that each method has its own application conditions. And we have also concluded that simpler methods allowing easier BEV operation, such as ICA-based and SOC-based methods, tend to have lower accuracy, while more complicated methods, like the data-driven method, with more difficult BEV operation tend to have higher accuracy. Therefore, hybrid methods are good compromises. For example, environmental factors which have great influences on ICA will result in a relatively substantial deviation. However, the data-driven method can use the environmental factors as the input characteristics for model training and link the factors with the run-in mechanism. Thus, a hybrid method of the ICA-based method and the data-driven method is very promising. Another example is a hybrid method of the SOC-based method and the data-driven method. This hybrid method uses the data-driven method to improve SOC prediction accuracy. Furthermore, a hybrid method of the model-based method and the data-based method can better determine model parameters.

8. Conclusion

This article gives a detailed analysis of SOH prediction for the LIBs. First, it introduces the LIBs and discusses the reasons for performance and degradation to make preparation for the SOH prediction. The definition and relationship between SOH and RUL is introduced. We then develop the framework for SOH prediction. Finally, the various methods are evaluated, and recommendations are proposed with actual industrial.

The internal reaction principle and aging mechanism of LIBs are complicated and describing them in specific models could be difficult. But there are currently numerous SOH prediction methods, with many having sound predictive capabilities in simulated environments but not in actual BEVs. The SOC and OCV are the most used methods in actual BEVs due to their simplicity and practicality. However, we consider the hybrid method, which combines the ICA and data-driven, to be more accurate and more

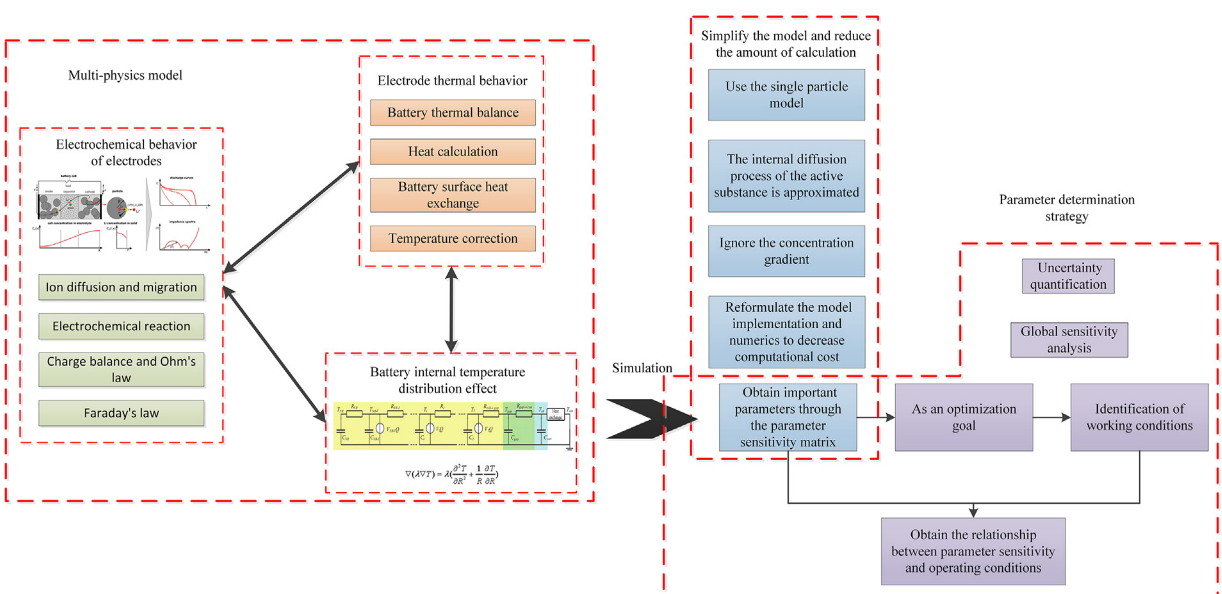


Fig. 25. The improvement and recommended strategies of SOH prediction based on the electrochemical model.

likely to be used in industrial production. The focus of future research is to make these SOH methods more accurate and apply these methods to predict actual BEVs.

Of course, although we try to present a comprehensive review of the SOH prediction, some aspects are not thoroughly explored. This article only introduces the SOH prediction research status in BEVs but not in HEVs and other NEVs. In addition, we also do not discuss the principle of LIBs, a more detailed aging mechanism, and the ways of delaying battery aging. At the same time, the prediction methods of SOH and RUL can also be introduced in more detail and the differences between them can be analyzed. In evaluating current SOH prediction methods, further research can be conducted by combining the actual industry. In addition to this, only a few methods are selected and provided improvement recommendations, but the SOH prediction still requires other various technical support, such as BMS, electrical, electronics, telecommunications, microcontroller technology, etc.

Declaration of competing interest

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

Acknowledgments

This work was supported by the National Natural Science Foundation of China (grant number: 61703406, 71602143, 51607122, 51806150), Tianjin Science and Technology Correspondent Project (grant number: 19JCTPJC47600), Tianjin Natural Science Foundation of China (grant number: 18JCYBJC22000, 18JCQNJC04400), and Tianjin University Innovation Team Training Program (grant number: TD130-5038).

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