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Review

Research Progress of Battery Life Prediction Methods Based on Physical Model

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Abstract: Remaining useful life prediction is of great significance for battery safety and maintenance. The remaining useful life prediction method, based on a physical model, has wide applicability and high prediction accuracy, which is the research hotspot of the next generation battery life prediction method. In this study, the prediction methods of battery life were compared and analyzed, and the prediction methods based on the physical model were summarized. The prediction methods were classified according to their different characteristics including the electrochemical model, equivalent circuit model, and empirical model. By analyzing the emphasis of electrochemical process simplification, different electrochemical models were classified including the P2D model, SP model, and electrochemical fusion model. The equivalent circuit model was divided into the Rint model, Thevenin model, PNGV model, and RC model for the change of electronic components in the model. According to the different mathematical expressions of constructing the empirical model, it can be divided into the exponential model, polynomial model, exponential and polynomial mixed model, and capacity degradation model. Through the collocation of different filtering methods, the different efficiency of the models is described in detail. The research progress of various prediction methods as well as the changes and characteristics of traditional models were compared and analyzed, and the future development of battery life prediction methods was prospected.

Keywords: lithium-ion battery; residual life; physical model; prediction method

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1. Introduction

Lithium-ion batteries have the advantages of high energy density, high output power, no pollution, no memory effect, small self-discharge, etc., and has become the main battery type of current new energy vehicles [1]. Common lithium-ion batteries on the market include lithium cobaltate, lithium manganate, and lithium iron carbonate batteries [2]. Lithium cobalt-acid batteries are the most widely used and have high specific energy, but the production of raw materials is more expensive; lithium manganate batteries have excellent multiplier performance and stability, but low energy density and poor safety [3]; lithium iron carbonate batteries have high energy density and good cycling performance, but poor low-temperature discharge performance [4]. There are many factors to measure the performance of the above lithium-ion batteries, and battery life is one of the key indicators and an important factor to be considered by users when purchasing electric vehicles [5]. During long-term use, the lithium-ion battery undergoes a series of electrochemical reactions and physical changes that degrade the performance and capacity until the end of its life [6].

State of health (SOH) estimation and remaining useful life (RUL) prediction are the basic problems of battery health management [7]. SOH represents the aging degree of lithium-ion batteries, which is mostly defined as the percentage of the capacity released by the battery from the full charge state to the cut-off voltage and the rated capacity of the battery under certain conditions [8]. RUL is a parameter that characterizes the deterioration of the battery to obtain the operating time from the beginning of the prediction to the end of the battery life [9]. The accurate prediction and estimation of SOH and RUL can effectively judge their future working capacity and identify problems in time to avoid unnecessary problems and losses [10].

RUL prediction methods based on the model from the perspective of the first principles are given a full explanation of the cell aging process, applicable to almost all of the conditions and operation mode. The analysis of a battery control strategy is also more detailed and accurate than other methods, which should be paid attention to and further developed in the process of RUL prediction research [11]. This paper will introduce the concepts related to RUL prediction, review the model-based RUL prediction methods of lithium-ion batteries, and make a comparison.

RUL refers to the number of charge point cycles a battery can perform before its life ends. There are many factors that affect the battery life, from the physical level including temperature, charging current, charging voltage, battery structure, etc. [12] and on the chemical level including the electrode material, electrolyte, battery resistance, etc. [13]. Generally, when the capacity of a battery is lower than 80% of the factory capacity, the battery is considered to be invalid. A common RUL definition is shown in Formula (1) below:

$$RUL = C_{EOL} - C_i \tag{1}$$

where C_{EOL} represents the number of charging and discharging cycles that can be carried out when the service life terminates, and C_i represents the number of charging and discharging cycles at the current moment [14].

In this paper, the existing model-based lithium-ion battery life prediction methods at home and abroad were divided into three categories: the electrochemical model-based method, equivalent circuit-based method, and empirical model based method [15,16], as shown in Figure 1.

Figure 1. Battery life prediction method based on the physical model.

2. Methods Based on Electrochemical Models

An electrochemical model is a model built by simulating the electrochemical reaction process of a battery [17]. It describes the laws of the cell from the point of view of internal physical and chemical processes including kinetic parameters, mass conversion processes, thermodynamic properties, mechanical, thermal, and electrical properties of materials [18]. It can provide a reference for battery research and development, so electrochemical models are often used for the analysis of battery principles and for battery research and development [19]. Electrochemical models include pseudo two-dimensional (P2D), single particle (SP) models, and coupled electrochemical models developed based on both.

2.1. P2D Model

The P2D model is shown in Figure 2. The battery was divided into three areas: negative electrode, diaphragm, and positive electrode [20]. Several governing equations were established in the P2D model to describe the diffusion of lithium ions in solid particles, mass transfer in the liquid phase, and the electrochemical reaction on the particle surface. Among these control equations, the most important is the Butler–Volmer kinetic response equation, as shown in Equation (2):

$$j = i_0 \left[exp\left(\frac{\alpha_a F}{RT} \eta \right) - exp\left(\frac{-\alpha_c F}{RT} \eta \right) \right] \quad (2)$$

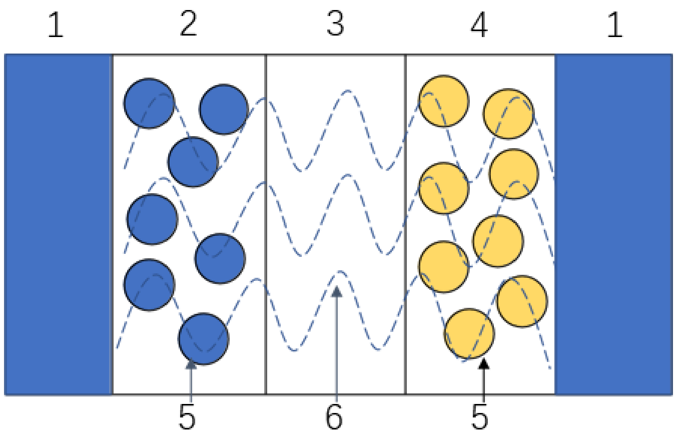

Figure 2. Structural diagram of the electrochemical model of a lithium-ion battery. (1) Liquid collector. (2). Negative electrode. (3) Diaphragm. (4) Positive electrode. (5) Active material. (6) Electrolyte.

In the formula, i_0 is the exchange current density, α_a is the anode transfer coefficient, usually taken as 0.5, F is the Faraday constant, R is the molar gas constant, T is the cell temperature, η is the spherical particle surface overpotential, and α_c is the cathode transfer coefficient, usually taken as 0.5.

The P2D model can deeply understand the mass transfer and kinetic reaction inside the battery, and lay a foundation for the degradation mechanism of the reactive battery and the optimal solution control of the battery [21]. In order to improve the prediction efficiency and accuracy of the model, researchers have optimized it from two aspects, one of which is to simplify the P2D model. Li et al. [22] used the finite difference method to discretely degrade the P2D model and obtained a simplified pseudo-two-dimensional model. On this basis, the electrochemical decay model (ADME) was proposed based on the decay aging phenomenon caused by the side reaction between the cathode and the anode. By comparing the experimental results, it was concluded that the model is simpler, and the maximum errors of predicting the terminal voltage variation trend under the hybrid pulse power characterization (HPPC) condition and 0.33 C constant current and constant power experiment were reduced by 3.92% and 3.94%, respectively. Compared with the simplified pseudo-two-dimensional model, the prediction accuracy was improved by 76% and 55%, respectively. Deng et al. [23] simplified the P2D model by polynomial approximation and proposed a polynomial approximation pseudo-two-dimensional (PP2D) model. The number of operations of this model was only one sixtieth of that of the P2D model. The calculation time of each step was less than 2 ms, which greatly improved the calculation efficiency. Another approach is to simplify electrolyte diffusion. Li et al. [24] combined the simplified electrolyte diffusion process and other kinetic processes of a lithium-ion battery, and a complete five-state diagonal system was obtained, which improved the prediction accuracy of the electrolyte concentration, electrolyte diffusion overpotential, and terminal voltage. All of these methods can simplify the P2D model. After obtaining the simplified model, the researchers optimized the parameter identification stage. Kim et al. [25] proposed a practical method to identify and select valid P2D model parameters, which changed significantly with battery aging. The average error of the P2D model output voltage between the experimental data and identification parameters was only 18.79 mV, which had high accuracy. Li et al. [26] used the heuristic algorithm for parameter identification, adopted a divide and conquer strategy, divided the P2D model parameter set into two groups, and identified each group of parameters separately. The complete identification of the P2D model can be completed within 10 h, which is 50% more efficient than the traditional identification method. Laue et al. parameterized the most prevalent electrochemical P2D model for Li-ion batteries [27]. A three-step technique was performed with quasi-static electrode measurements of open-circuit potentials, C-rate testing, and electrochemical impedance spectroscopy all taking place. Each step's identifiability was thoroughly addressed, and basic guidelines for future parameterizations were generated. According to the findings, open-circuit potentials and C-rate measurements are insufficient to properly parameterize electrochemical models. To address the ambiguity of diffusion and electrical processes in quasi-static circumstances, highly dynamic tests such as impedance spectroscopy are required. The findings of this study offer recommendations for the usage of electrochemical models in applied science and industry. To parameterize the most often used electrochemical pseudo-two-dimensional model, Xu et al. [28] suggested a unique nondestructive parameter identification approach. First, the sensitivity of the model parameters was examined and divided into three groups based on the situations under which the parameters were most sensitive. Second, for these unknown values, a deep learning technique was utilized to provide plausible first predictions. Finally, two alternative approaches for parameter identification were coupled in order to progressively estimate the parameters with great sensitivity. The results indicate that utilizing both the simulation and experimental data, one electrochemical parameter could be properly calculated in 14 h. The root-mean-square error of the model forecast voltage was less than 14 mV after evaluating the model parameters.

2.2. SP Model

Compared with other electrochemical models, the single-particle model is widely used because of its simple construction and fast prediction speed [29]. In the literature, the SP model was simplified to degrade to improve the calculation speed and prediction accuracy of the model [30,31]. At the same time, various factors that may affect the battery were taken into account in the model, and internal indicators were selectively extracted to track the battery health status [32,33]. Sadabadi et al. [34] proposed a RUL prediction algorithm based on single particle model parameter estimation. The algorithm uses the moles of circulating lithium and battery resistance as indicators of the state of health, and uses the derived composite SOH metric to design particle filter-based RUL predictions [35], where the greater the SOH-related estimates, the more accurate the RUL estimate. Based on the SP model, Li et al. [36] proposed an improved simplified model, obtained physical parameters with the aging mechanism, and selected new health indicators by analyzing the relationship between the physical parameters and battery health status [37], which is helpful in accurately predicting the remaining life of the battery, and has important theoretical significance and practical value for improving the level of battery management technology. Prasad et al. [38] simplified the model by two key parameters, battery resistance, and cathode solid-phase diffusion time [39]. The model uses the gradient parameter real-time update method under the excitation of the HPPC current [40]. Within 200 s, the estimated value of the total battery resistance and diffusion time could converge to the optimal value within 99%. The calculation speed was fast, and can be used for the online estimation of the remaining life of the car battery. Deng et al. [23] employed a sequence of polynomial functions to simulate the electrolyte phase concentration profile, the solid phase concentration profile, and the nonuniform reaction flow profile, respectively, to simplify the P2D model. The accuracy of the second- and third-order polynomial estimates for the reaction fluxes was compared, and the higher order's more robust correctness was proven. The created model was simulated using a variety of constant flow rates, mixing pulses, and driving times, and the results were compared to the P2D model and the original SP model. The findings revealed that the proposed model captures the cell properties adequately while significantly reducing the computational complexity.

2.3. Electrochemical Coupling Model

The above electrochemical models can be optimized in their respective fields to compensate for deficiencies to a certain extent, but some deficiencies are inherent to the models themselves and cannot be eliminated. In this case, the integration with other models is needed to improve the accuracy. The P2D model is an isothermal model without considering the coupling relationship between heat generation and chemical reaction. In the actual charging and discharging process, the battery will inevitably generate heat. Kuangke et al. [41] put forward a kind of large capacity motor such as the lithium-ion battery electrochemistry-thermal coupling efficient modeling method through the classification of the parameters, and the measured/identification parameters for precise measurement and parameter identification, using pulse charging experiments under different temperature calibration solid phase diffusion coefficient of the DS and reaction rate constant k, further established the battery heat production model. The simulation data of the voltage and temperature agreed well with the test data, and the parameters of the model were more accurate at room temperature. The average voltage error was less than 10 mV, and the average temperature error was less than 1.1 °C. The established model had good accuracy and adaptability. Wang et al. [42] also developed an electrochemical-thermal model utilizing P2D and detailed how some parameters varied with temperature. The proportional mean square variance between the model's anticipated and observed residual life was only 0.625%, which compensated for the simulation error caused by the temperature change. Zhang et al. [43] discovered that the P2D model's impedance parameters were only consistent with the actual battery at low frequencies. It obtained high simulation precision in the wide bandwidth range from 10 mHz to 1 kHz by integrating the P2D model with the EIS model. Under the sinusoidal electrical excitation of 5 Hz, 10 Hz, and 20 Hz, the root mean square error of the improved model decreased by 24.8%, 30.6%, and 33.0%, respectively, when compared to the P2D model. Li et al. [36] found that the existing SP model did not consider the degradation mechanism, and proposed a new prediction model combining the capacity degradation model and SP model. This model could quickly predict the capacity attenuation and voltage distribution changes with the number of cycles and temperature. Compared with the experimental results, the root mean square error of the model prediction was 0.0103. A coupled electrochemical-thermal coupling (ECT) model of LiCoO batteries was proposed by Li et al. [44] to describe their charging and discharging behavior. Calculations of heat generation, conduction, and dissipation were added to the simplified electrochemical model by means of a total set thermal analysis and a rational reduction and recombination of the cell mechanism parameters to reduce the estimation complexity. Specifically designed identification conditions were used to obtain the mechanism parameters based on the excitation response analysis. The applicability of the model under different operating conditions was verified by simulations. The simulation results of the end voltage and surface temperature were in good agreement with the actual experimental measurements at lower C-rates and dynamic load currents. Jiang et al. [20] created a one-dimensional (1D) electricity generation-three-dimensional (3D) thermal connection model to investigate the heat transfer mechanism of hexagonal Li-ion cells cooled on various exterior surfaces. The simulation parameters studied were the forced convection cooling coefficient, h, the thermal diffusion surface area, and the dimension of the cell. The variation in temperature of the prismatic cells with conduction force cooling on tiny side surfaces was found to be more uniform than that of prismatic cells with massive front area cooling. The highest temperature differential of the prismatic cell with tiny side surface cooling was kept constant at h = 100 W/m^2 K as the cell size rose. Furthermore, the influence of operating temperature on the capacity degradation of Li-ion batteries during cycling was examined. The greater the operating temperature, the faster the parasitic lithium/solvent reduction process, which resulted in the depletion of lithium ions and enhanced the rate of capacity degradation throughout cycling.

2.4. Comparison of Electrochemical Models

In order to more clearly show the advantages and disadvantages of the prediction methods based on electrochemical models, the prediction methods are summarized. In this paper, "★" was used to indicate the accuracy and complexity of the prediction methods. The results are shown in Table 1.

Table 1. Electrochemical model.

Model	Advantages	Disadvantages	Prediction Accuracy	Complexity
P2D model	It can describe the internal dynamic behavior of the battery, and has the advantages of accurate model and high calculation accuracy.	There are many parameters in the model and the calculation is complicated and the efficiency is low.	High ★★★★☆	High ★★★★☆
SP model	The modeling complexity is low and the calculation accuracy is high.	The physical properties of electrolyte are ignored and the problem of order reduction is not considered.	Higher ★★★★★	Lower ★★☆☆☆
Electrochemical fusion model	Strong robustness and high prediction accuracy.	The model is complicated due to the large amount of calculation and many optimization parameters.	High ★★★★☆	High ★★★★☆

3. Method Based on Equivalent Circuit Model

The equivalent circuit model is a kind of model that expresses the external characteristics of the battery through the combination of circuit components. The relationship between the various influencing factors of the battery and the state of charge can be given, so it is widely used. This mainly includes the Rint model, Thevenin model, PNGV model, and RC model. The RC model was divided into the integer model and fractional model.

3.1. Rint Model

The Rint model equates the battery to a series of the ideal voltage sources and the internal resistance, which is the simplest equivalent circuit model. Resistors were used to simulate the ohmic and polarizing internal resistance of batteries. The circuit structure is shown in Figure 3.

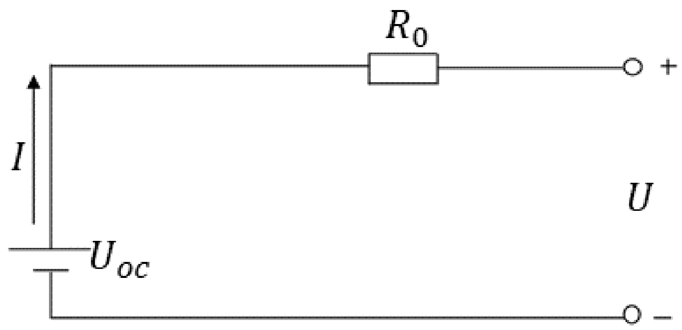

Figure 3. The Rint circuit structure diagram.

The structure is simple and the parameters are easy to calculate. However, it cannot describe the dynamic process and is seldom used in practical applications [45]. Researchers have improved the accuracy by connecting several circuit models in series and parallel [46].

3.2. Thevenin Model

The Thevenin model considers the polarization phenomenon in the battery reaction, and uses the parallel link of resistance and capacitance to simulate the complex internal reaction of the battery in the charging and discharging process [47] (see Figure 4).

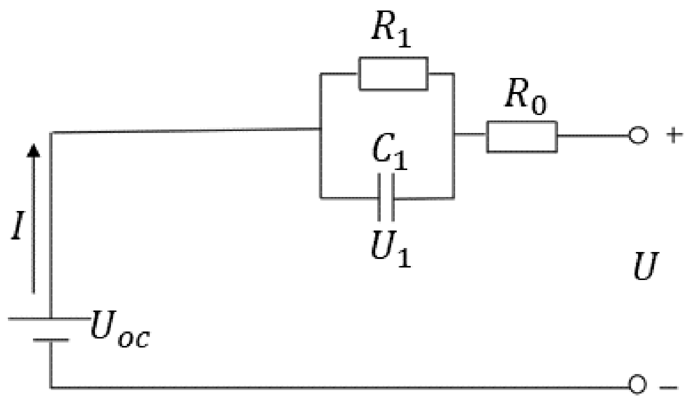

Figure 4. The Thevenin circuit structure diagram.

In the model, the ideal voltage source describes the open circuit voltage of the battery, the resistance is the ohm internal resistance of the battery, and the parallel connection of capacitance and resistance describes the polarization of the battery [48]. The Thevenin equivalent circuit model can simulate the dynamic and static characteristics of the battery very well, taking into account the nonlinearity of the battery [49], and the parameters are obtained in a simple way that can be easily translated into a state space model [50–52]. However, in the actual operating environment, some model parameters of the battery may change with environmental conditions such as temperature. Hossain [53] proposed a temperature compensation model parameter extraction method based on the Thevenin model was proposed, which can accurately extract the battery parameters within the operating temperature range of −5~45 °C. Nikohan et al. [54] proposed a smoothing and adaptive adjustment method for parameter estimation, which enabled the model to have good prediction accuracy under different temperatures and load profiles. Ding et al. [55] proposed improving the Thevenin model by taking temperature into account when calculating battery voltages in open circuits for lithium-ion batteries. The precision of the battery connection voltage computation was increased without expanding the model's order. The model was suggested based on the Thevenin model and the association between the open-circuit voltage and charge state; then, the battery model parameters were determined using polynomial fitting and the genetic algorithm, correspondingly, based on the findings from the experiments of the open-circuit voltage test and the combined power pulse characteristic test. The suggested model was evaluated and verified under dynamic stress test settings at various temperatures and driving schedules on an urban multimeter. The suggested model was very accurate, with less than 1% error in parameter identification. Lyu et al. [56] suggested a prototype-based and driven by data technique to estimate the lithium-ion battery health. To test the resilience of the stated aging feature, an independent experimental design and a two-way ANOVA were utilized. The Box–Cox conversion was implemented to improve the consistency of the aging parameters at low temperatures when looking at the influence of temperature on battery performance. Experiments on the battery lifespan validated the estimate efficacy of the suggested strategy. The experimental findings demonstrated that the suggested approach had good estimation precision at various temperatures. Back propagating networks and support vector regression techniques were used with the same aging characteristics to test the estimate framework's universality. Sun et al. [57] refined the equivalent ohmic resistance in the Thevenin model and replaced the original ohmic resistance R_0 with R_{chg} when the battery is charged and R_{dis} when the battery is discharged. The improved Thevenin model is shown in Figure 5. The improved Thevenin model consisted of three main components including the open-circuit voltage U_{oc}, the internal resistance, and the equivalent capacitance. The internal resistance consists of the ohmic resistance R_0 (including R_{chg} and R_{dis}) and the polarization resistance R_p. The equivalent capacitance C_p is used to describe the transient response during charging and discharging. Rise is the voltage across C_p and I_P is the outflow current from C_p. Equations (3) and (4) are as follows:

$$U_t = U_{oc} - U_p - i_L R_0 \tag{3}$$

$$\dot{U_p} = \frac{i_L}{C_p} - \frac{U_p}{C_p R_p} \tag{4}$$

where i_L is the load current, which is positive when discharging and negative when charging, and Ut is the terminal voltage.

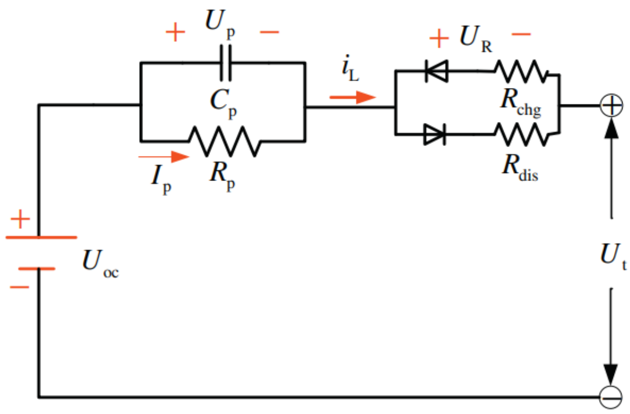

Figure 5. Schematic diagram of the improved Thevenin battery model.

3.3. PNGV Model

The PNGV model is based on Thevenin's model with a capacitor C_0 in series to describe the change in battery open-circuit voltage due to the accumulation of load current over time (see Figure 6).

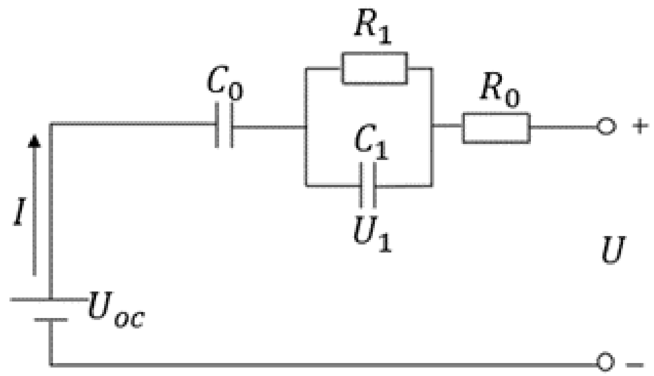

Figure 6. The PNGV circuit structure diagram.

Its significant feature is that the capacitance is used to describe the change characteristics of the open circuit voltage generated with the time accumulation of the load current when the battery absorbs and emits electricity. The capacitance reflects the capacity of the battery. However, the disadvantages are also obvious. The model cannot well-describe the ground polarization characteristics of the battery and it is difficult to identify the battery online. To solve these problems, researchers have obtained the improved PNGV model by connecting a RC parallel circuit in series to the PNGV model to represent the polarization reaction, as shown in Figure 7.

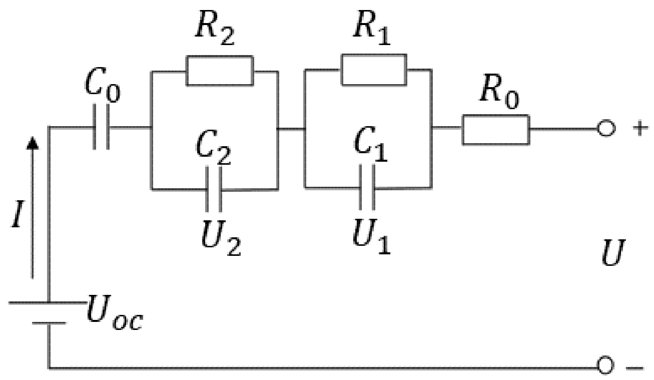

Figure 7. The improved PNGV circuit structure diagram.

The RC parallel circuit is composed of polarization resistance R1 and polarization capacitance C1. Compared with the off-line estimation, the maximum error of this method was reduced by 35.106%, and the root mean square error was reduced by 25.38%, which improved the prediction accuracy of the mode [58–60]. Lin [61] et al. transformed the PNGV model into a difference equation that could be identified online by the discrete method, laying a solid foundation for various state estimations. Liu et al. [62] established a partnership of new generation vehicle (PNGV) battery models for the inaccuracy of state of charge (SOC) estimation and proposed an open circuit voltage method. The model parameters for different SOC states were calculated by the charging and discharging process of the Li-ion battery. Depending on the PNGV model, both the internal resistance compensation technique and the Kalman filter approach are suggested. On this basis, simulations and experiments were conducted for both methods to obtain the open circuit voltage and battery SOC. The experimental findings revealed that the PNGV model is reliable and can represent the battery's properties during the discharge operation. The ampere–hour (or current integration) method's accumulated inaccuracy and inaccurate measurement of the beginning value can be prevented by correctly applying the SOC calculation. Furthermore, the estimated SOC may be kept extremely exact. Gao et al. [63] created a PNGV-based battery model for immediate form parameter recognition based on the characterization of LiFePO4 Li-ion batteries. The gradual memory distribution recurrent least squares approach was used to identify the PNGV model parameters in real-time. The simulation findings suggest that the approach can more successfully discover model parameters in real-time and decrease the inaccuracy of the data overload recursive least squares method. It may be used to improve the accuracy of the SOC estimate for LiFePO4 Li-ion batteries. Yuyang et al. [64] suggested a novel verification model that takes into account variances in battery charging and discharging and used an organic light emitting-resistance parallel circuit instead of the usual PNGV model's inner resistance. To assess the changing and static characteristics of the battery, a resistor-capacitor (RC) parallel network was utilized. The tripartite lithium-ion battery was investigated, and the improved model was employed for online parameter recognition by employing the forgetting component recursive least squares approach. The primary charge and discharge experiments were described in order to mimic and assess the lithium battery's operational properties. The experimental findings suggest that the modified 2RC-PNGV model may more accurately depict the Li-ion battery's operational characteristics. The HPPC experiment's mean voltage error was 0.17%, and the model was quite accurate. During the primary charging phase, the typical error of SOC estimation was 0.957%, with a maximum estimate error of 5.03%. The average error of SOC estimation for the primary discharge process was 0.807%, with an elevated estimation error of 3.38%. The findings demonstrate that the SOC can be estimated using both the revised 2RC-PNGV model and the joint estimation technique.

3.4. RC Model

In practical applications, the equivalent circuit model is generally used as the battery model, which needs to be selected according to the chemical characteristics of the battery and the computing capability of the processor. The accuracy of first-order RC is lower [65–67], while high-order (third-order and above) models are cumbersome in calculation and have a general effect on improving the accuracy. The second-order RC equivalent circuit model can describe the actual terminal voltage of the battery well, and the complexity of the model was low (see Figure 8).

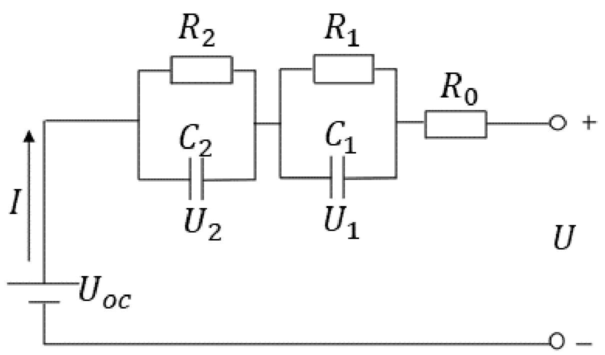

Figure 8. Second-order RC circuit structure diagram.

The second-order RC model is often combined with algorithms in battery life estimation such as the differential evolution (DE) algorithm, extended Kalman algorithm (EKF) algorithm, adaptive genetic algorithm (AGA), which can improve the accuracy and speed of prediction [68–71]. Guha et al. [72] proposed a new parameter estimation method for fractional-order battery model by using recursive least squares (RLS) combined with fractional-order state variable filters (FOSVF). The resistance model developed from the predicted parameters is capable of reconstructing the EIS spectra of the genuine lithium-ion battery with high accuracy. Furthermore, the impacts of aging on the battery metrics and EIS characteristics were investigated. Nejad et al. [73] provided a sensitivity analysis of two time-constant RC circuit models for the resistive-capacitive elements they formed, and determined the model parameters of a cylindrical lithium iron phosphate battery using electrochemical impedance spectroscopy techniques and nonlinear least squares. The results show that the model parameters varied with the state of charge of the battery, and the importance of each parameter for calculating the average available power (power state) of the battery in a given frequency band was analyzed. Ji et al. [74] investigated the estimate and modeling of lithium-ion battery state of charge (SOC). The ampere–hour (Ah) incorporation approach based on external features was investigated as well as the open-circuit voltage (OCV) technique, to integrate the two approaches to determine SOC. Given the model's precision and intricacy, a second-order RC analog circuit model of the Li-ion battery was adopted. The lithium-ion battery's associated properties were derived using pulsing discharge and exponential matching. Fixed-resistance capacitance and variable-resistance capacitors were used in the experiments. The variable-resistance capacitor model had an accuracy of 2.9%, proving the validity of the suggested model.

3.5. Comparison of Equivalent Circuit Models

In order to more clearly show the advantages and disadvantages of prediction methods based on equivalent circuit model, the prediction methods are summarized, and the results are shown in Table 2.

Table 2. Equivalent circuit model.

Model	Advantages	Disadvantages	Prediction Accuracy	Complexity
Rint model	The model is simple and the parameter calculation is simple.	Unable to describe dynamic processes, poor accuracy when using high current, ignoring battery characteristics.	Lower ★★☆☆☆	Lower ★★☆☆☆
Thevenin model	In practical engineering applications, the polarization effect and battery characteristics are considered.	The stability of the model is poor, and factors such as battery aging and temperature change have great influence on the accuracy of the model.	Medium ★★★☆☆	Medium ★★★☆☆
PNGV model	Considering the influence of temperature, the model is robust and accurate.	The cumulative error of series capacitance will reduce the model accuracy and cannot reflect the polarization phenomenon well.	High ★★★★☆	Medium ★★★☆☆
RC model	The calculation is moderate and the model has high precision, which is closer to the real battery characteristics.	The calculation of structure and parameters is complicated.	High ★★★★☆	High ★★★★☆

4. An Empirical Model-Based Approach

The empirical model-based RUL prediction method builds the degradation model by fitting the historical degradation data of lithium ion battery with the empirical model, and updates the model parameters by the filtering method [75]. Finally, the battery life prediction is realized [76]. This model does not need to analyze the internal electrochemical reaction and has a wider application range [77]. The empirical models mainly include the exponential model, polynomial model, multi-mixture model, and capacity regeneration model, and the filtering methods mainly include Kalman filtering (KF), particle filtering (PF), and their improved algorithms [78].

4.1. Exponential Model

The mathematical formula used to construct the empirical model in this model is the first single exponential formula, as shown in Formula (5):

$$C_k = a_1 exp(a_2 k) \tag{5}$$

where C_k is the capacity of the battery at the KTH cycle and a_1 and a_2 are the model parameters. This empirical model made some improvements in the RUL prediction and attracted the attention of some researchers [79,80]. However, this model classifies the very short battery rest time as the capacity regeneration period, leading to a large error in prediction. The examination of a vast number of findings from the experiments revealed that the battery capacity deterioration is directly connected to a rise in internal resistance, which can be described as an exponentially increasing sum. The experiments and experience revealed that the dual exponential empirical decline model fit the nonlinear battery capacity deterioration well [81], then the double exponential formula was introduced as shown in Formula (6):

$$C_k = a_1 exp(a_2 k) + a_3 exp(a_4 k) \tag{6}$$

The number of model parameters was raised to four. Qin et al. [82] used the particle filter PF method as the foundation and the two-dimensional exponential model as the state solution. The deterioration curve of the battery was established after continuous sampling, and the remaining service life was assessed. The experimental findings demonstrated that the model has a strong prediction effect and can effectively fit the battery deterioration curve, and the forecast accuracy will steadily improve as the cycle increases. Zhang et al. [83] devised a technique for predicting residual usable life (RUL) based on an exponential model and particulate filtering, which takes into account the nonlinear and non-Gaussian capacity decline features of lithium batteries. The prediction performance was evaluated using the prognostic horizon index and the new specific accuracy index. In addition, the prediction errors under different prediction starting points were given. The suggested methodology outperformed methods such as the integrated autoregressive moving average model, the merged nonlinear deficient self-regressive model, and the established particle filter algorithm in terms of prediction performance. The suggested prediction approach exhibited a higher prediction accuracy and integration according to the precision index. The RUL prediction of lithium batteries can help maintenance and support systems make better decisions in order to improve the maintenance methods and save on maintenance costs. Ma et al. [84] suggested a particle filter-based enhanced exponential model for a data-driven method to lithium-ion battery remaining lifespan. Four case studies were conducted to validate the suggested forecast method's excellent accuracy in predicting and low uncertainty. Using the particle filtering approach, we compared the residual service life forecast findings linked to the original exponentially model. The experimental findings revealed that the revised exponential model needed less parameters than the original model; the suggested prognostic approach had a steady and high prediction accuracy, and the suggested approach had a low level of uncertainty. Yang et al. [85] suggested a Bayesian model-based technique to predict the remaining usable life (RUL) of various sorts of batteries. First, two logit models were created to represent the battery deterioration pattern. It was empirically proven that the new model surpassed the previous empirical cell deterioration models by fitting the data. A particle filter-based prediction approach was subsequently incorporated into the model to forecast the battery's potential deterioration trajectory. The findings revealed that the suggested prognostic technique outperformed the two existing exponential models in terms of the prediction accuracy. Tseng et al. [86] used statistical approaches to create a regression model for battery prediction. The resultant regression framework not only analyzed the battery's deterioration trend, but also estimated its remaining usable life (RUL) at an early stage. To obtain the optimal regression model parameters, a particle swarm optimization (PSO) approach was used. The simulation findings suggest that the regression models utilized as aging parameters can produce more accurate health status profiles than the count of cycles.

4.2. Polynomial Model

The exponential model obtained the best fit in the nonlinear stage of capacity degradation, while the polynomial model obtained the best fit in the linear stage of capacity degradation [87–89]. Formula (7) is shown below:

$$C_k = a_1 k^3 + a_2 k^2 + a^3 k + a_4 \tag{7}$$

Sun et al. [90] applied a third-order polynomial model to fit the battery health degradation process, applied particle filter algorithm to predict the remaining service life of the battery, and gave the probability density function of the remaining service life of the battery. The health degradation model and the battery remaining life prediction were verified using examples. Su et al. [78] improved the original polynomial model, so that the model had fewer parameters and was more suitable for the PF prediction method. The cell was modeled using a polynomial model by Azis et al. [91]. A generalized lowered gradient (GRG) optimization approach was then used to forecast the parameters. A double elongated Kalman filter (DEKF) approach was then utilized to estimate both the SOC and SOH. The DEKF technique findings were compared to the EKF method to determine the estimator's performance. The numerical results indicate that the DEKF approach accurately calculated the battery's SOC, internal resistance, and volume. The DEKF also provided superior results when estimating the battery's SOC. The nonlinear relationship between the circuit parameters and SOC was explicitly described using an analytic polynomial function by Wang et al. [92]. The influence of the polynomial order was explored systematically through fitting and prediction accuracy, with the leave-one-out cross-validation (LOOCV) method employed to assess the prediction performance. The EIS measurements were performed on a 20 A–h commercial lithium battery to verify the validity of the proposed model. The results showed that the seventh-order polynomial function is sufficient to capture the nonlinear effects of SOC on the circuit parameters.

4.3. Exponential and Polynomial Mixed Model

The exponential and polynomial mixed model is an integrated model that combines an exponential model and a polynomial model, which takes into account the global and local regression characteristics, and is a more accurate parametric model than the two single models [86]. Because model parameters fluctuate with the dynamic features of battery degradation, it is preferable to utilize PF to estimate and adapt model variables to track the battery deterioration process with quadratic and non-Gaussian properties. Formula (8) is shown below:

$$C_k = a_1 exp(a_2 k) + a_3 k^2 + a_4 \tag{8}$$

Xing et al. [93] tracked the degradation trend of the battery during the cycle life based on a multi-finger model, and used the PF method to adjust the model parameters online. The prediction performance of the ensemble model, the exponential model, and the polynomial model was compared through experiments, and it was proven that the exponential model had strong robustness. A new interaction-based multi-model particle filtering (IMMPF) data-driven prediction method for determining the remaining useful life (RUL) of lithium-ion (Li-ion) batteries and the probability distribution function (PDF) of the uncertainty associated with the RUL was proposed by Wang et al. [94]. The IMMPF was used for various state problems. Models of battery capacity deterioration are useful in predicting the RUL of Li-ion batteries. Using three enhanced models, the IMMPF approach was used to calculate the RUL of Li-ion batteries. The experimental results revealed that the one-dimensional formula of state particle filter (PF) is better suited to predicting the long-term trend of the capacity of batteries. The suggested technique, which involves the collaboration of various models, has demonstrated stability and good prediction accuracy as well as the capacity to reduce the uncertainty of forecasting the RUL PDF for lithium-ion batteries. Gou et al. [95] introduced a combined RUL prediction approach, ORV-MDMHD, which combined the optimum association vector (RV) with an enhanced degradation model (MDM) and Hausdorff distance (HD). To improve the long-term impact of ability data on the forecast, a phase space restoration was employed to generate inputs to the RVM; the curve resemblance metric HD was used to choose the best fit that most closely resembled the actual deterioration curve. The anticipated RUL of the battery may be computed by extending the optimum curve to the failure threshold. The experimental findings on the two battery examples demonstrate that the suggested prediction approach may provide stabler forecasts and greater accuracy, particularly for the long-term prediction of the lithium battery RUL.

4.4. Capacity Regeneration Model

Lithium-ion battery data have the problem of capacity regeneration. The longer the cycle interval, the more obvious the capacity regeneration phenomenon. Describing the problem of capacity regeneration in a model remains a huge challenge. In general, the capacity loss of the battery from the first cycle to the Nth cycle is shown in Formula (9):

$$Q_{loss}^{(N)} = S_{neg} \sum_{n=1}^{N} \int_0^{t_{cc,n}} J_s^{(n)} dt \quad (9)$$

In the above formula, $J_s^{(n)}$ represents the density of the side reaction currents of the nth cycle, $t_{cc,n}$ represents the charging time in the nth cycle's continuous current stage, and S_{neg} represents the total interface area of the negative electrode—the side reaction current density of the first cycle. Guha et al. [96] combined the battery capacity degradation model based on the battery capacity test data and the internal resistance growth empirical model based on the EIS test data, and used it in the PF framework to obtain a new remaining service life estimation method, which improved the prediction accuracy. Pan et al. [97] mixed particle filter, exponential smoothing, and the capacity degradation model to obtain a new prediction method, which had a higher accuracy and stability than the pure particle filter method. Such estimation methods, combined with filtering algorithms, have gradually become the mainstream [98]. Pang et al. [99] introduced a new approach for estimating the RUL of lithium-ion batteries that combined a wavelet breakdown technique (WDT) with the nonlinear autoregressive neural network (NARNN) model. First, the multiscale WDT was used to distinguish between the global decline and local renewal of the battery capacity sequence; next, a RUL prediction architecture based on the NARNN model was built for the recovered global degradation and local renewal. Finally, the two prediction result portions were merged to obtain the final RUL prediction results. The experiments demonstrate that the suggested technique not only captured the capacity renewal phenomenon successfully, but it also had high forecast accuracy and was less impacted by varied prediction starting points. Ma et al. [100] used a combination of particle filtering (PF) and the Mann–Whitney U test (PF-U) to determine the capacity renewal point (CRP). For the RUL prediction, the autoregressive (AR) model and PF algorithm were utilized. The PF algorithm's deteriorated model parameters were updated utilizing the capacity projected by the AR model, and the method's efficacy was validated using the NASA Li-ion battery dataset. As a result, the technique provided in this study has the best accuracy, offers a platform for detecting the point of capacity regeneration, and minimizes the RUL error in forecasting even more.

4.5. Comparison of Empirical Models

In order to more clearly show the advantages and disadvantages of the prediction methods based on the empirical model, the prediction methods are summarized, and the results are shown in Table 3.

Table 3. Empirical model.

Classification	Advantages	Disadvantages
Exponential model	For the nonlinear stage of capacity degradation, the fitting degree is high.	The linear stage of capacity degradation is poorly treated.
Polynomial model	The fitting degree is high for the linear stage of capacity degradation.	The nonlinear stage of capacity degradation is poorly treated.
A hybrid exponential and polynomial model	High accuracy and strong robustness.	Complex structure and many parameters.
Capacity degradation model	Besides the charge and discharge state of the battery, the rest state of the battery is also considered.	Insensitive to capacity degradation and regeneration.

After the above model is constructed, the model parameters need to be updated by the filtering method, and finally, the battery life prediction is realized. Here is a brief introduction of the filtering method.

4.5.1. KF Algorithm

Kalman filtering [101] is a linear optimal state estimation method, which is known as one of the most famous Bayesian filtering theories. The equation of state is a linear representation of w_k, u_{k-1} and x_{k-1}.The observation equation is a linear representation of x_k and v_k. The state equation of the Kalman filter is shown in Formula (10):

$$x_k = Ax_{k-1} + Bu_{k-1} + w_k \tag{10}$$

The observation equation is shown in Formula (11):

$$z_k = Hx_k + v_k \tag{11}$$

In the above formula, x_k represents the state vector, z_k represents the observation vector, A represents the state transition matrix, H represents the observation matrix, w_k represents the system noise vector, u_{k-1} represents the system control vector, and v_k represents the observation noise vector.

There are relatively few studies on RUL prediction based on KF, because lithium-ion batteries have strong nonlinear non-Gaussian characteristics, and KF is not suitable to deal with this kind of problem [84], so PF is the focus of research in this field. Models are mostly combined with PF to increase the budget accuracy.

4.5.2. PF Algorithm

The process of resembling the probability density function by seeking a collection of random specimens propagating in the state distance, and replacing an integral operation with the sample implies this; then, getting the smallest possible variance estimates of the system state is referred to as the particle filter. These samples are referred to as "particles" visually, thus the name particle filter [102]. The equation is shown in Formula (12):

$$P(x_t|z_{1:t}) \approx P_N(x_t|z_{1:t}) = \sum_{i=1}^{N} w_t^i \delta\left(x_t - x_t^i\right) \tag{12}$$

In the above formula, x_t^i represents the state value of the ith particle at time t, $z_{1:t}$ represents the observation value at time 1~t, w_t^i represents the weight of particle i at time t, and δ represents the Dirac function.

The superiority of particle filter in nonlinear and non-Gaussian system determines its wide application in the battery life field.

5. Conclusions

In this paper, we systematically reviewed the current research status of power battery remaining life prediction, compared and analyzed the advantages and disadvantages of existing battery models and prediction methods, and summarized the future research and development trends as follows:

(1) To improve the efficiency of electrochemical model prediction, current research focuses on simplifying the model while considering as many factors as possible and determining the parameters by different methods. In addition, with the continuous development of modern technology, health factors are no longer limited to traditional parameters such as voltage and current. In the future, some new factors can be extracted by ultrasonic and infrared technologies to meet the requirements of small number and comprehensive reflection, thus improving the accuracy and range of prediction.

(2) The simulation accuracy of a single equivalent circuit is low, so series resistors or capacitors were used to improve the dynamic stability during the study, which reduced the

influence of environmental factors and incorporated parameter identification into the algorithm to compensate for the poor prediction accuracy. However, as the number of series-connected components increased, the cumulative error also increased. How to reduce this error is one of the focuses of future research on equivalent circuit models.

(3) The empirical model-based RUL prediction method constructed a degradation model by fitting the historical degradation data of lithium-ion batteries with an empirical model, and used a filtering method to update the model parameters to achieve the RUL prediction of batteries. The simple empirical model established the relationship between the battery characteristics through complex mathematical formulas, which had a low prediction accuracy and poor stability. Based on this, PF, KF, and their improved filtering methods were used to update the data, and influence factors were added to improve the accuracy and reduce the error. Future research will focus on finding more comprehensive mathematical methods to construct empirical models and update the model data by other intelligent optimization methods.

From the current research status of the remaining life prediction methods, combining optimization algorithms with battery models has become the mainstream method. However, there are some problems with this method such as how to design an effective method to predict the lifetime of lithium batteries in multiple scenarios and improve the accuracy of the prediction, which is still a challenge. In order to improve the accuracy of this method's prediction, researchers should actively explore more pervasive parameter optimization algorithms. With the rapid development of Internet technology and artificial intelligence, it is believed that breakthrough progress will be made in the life prediction technology of lithium-ion batteries in the near future.

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