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# An electrochemical model based degradation state identification method of Lithium-ion battery for all-climate electric vehicles application

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## HIGHLIGHTS

- The electrochemical model has been proposed in this study.
- The finite analysis method and numerical computation method are used to solve PDEs.
- Five aging characteristic parameters are extracted to describe health state LiBs.
- The degradation trajectories of LiBs at different temperatures are built.
- An HIL test is conducted to verify the accuracy of the electrochemical model.

## ARTICLE INFO

*Keywords:*
All-climate electric vehicles
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Electrochemical model
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## ABSTRACT

The Lithium-ion batteries (LiBs) are the core component of the all-climate electric vehicles. The aging state recognition is carried out based on the proposed electrochemical model (EM) instead of the traditional equivalent circuit model (ECM) and black boxes model in this paper. Firstly, a group of mathematical equations are built to describe the physical and chemical behaviors of batteries based on the electrochemical theory. Then, the finite analysis method and the numerical computation method are used to solve the mathematical equations and the model has been built. Next, the optimization algorithm is used for identifying the parameters of the model. The aging state recognition of the battery on whole lifetime is carrying out based on the ageing data. Five aging characteristic parameters are determined to describe the health state of the battery, and their degradation trajectories are obtained. Finally, a battery-in-loop approach is employed to verify the model based degradation recognition. Results show that the maximum voltage error is within 50 mV and the state of health estimation error is bounded to 3%.

## 1. Introduction

Due to the urgency of environmental pollution and energy shortage, replacing the traditional fuel vehicles by electric vehicles (EVs) gradually has become a global consensus [1,2]. With the large-scale commercialization of EVs, how to solve the problem of EV's usage in the cold environment has been put on the agenda. EVs will be used as the major means of transportation in 2022 winter Olympic Games, when the weather is quite cold in Beijing. The state of health (SOH) represents the aging state of the battery. Accurate and reliable SOH estimation is particularly important for the safety of LiB safety management in all-climate EVs because low temperature will accelerate the battery aging, which might lead to a series of security issues [3]. The reasons of the battery aging can be attributed to the large number of side effects during the usage of the battery, resulting in the internal electrode materials decomposition and electrolyte conduction ability decline. Therefore, in order to evaluate the health state of LiB, it is necessary to extract the physical and chemical parameters of the battery and establish the relationship between the relevant characteristic parameters and battery aging.

SOH estimation methods can be divided into experiment-based methods and model-based methods. The experiment-based method is simple and convenient, but a large number of experiments are required to obtain the resistance and capacity change during the aging process. The authors in [4,5] used electrochemical impedance spectroscopy (EIS) as a situ analysis tool to observe the aging degree under different operating conditions and identified the aging parameters with EIS technique. The model-based approach, based on the battery model,

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**Nomenclature**

| | |
|---|---|
| $\varepsilon$ | active material volume fraction |
| $c$ | lithium ions concentration |
| $D$ | diffusion coefficient in electrode |
| $a$ | specific surface area of electrode |
| $j$ | molar flux of lithium ions |
| $t_+^0$ | Li-ions transfer number in the electrolyte |
| $r$ | coordinate along the radius of active particles |
| $x$ | coordinate along the electrode |
| $R_s$ | radius of spherical active particle |
| $\kappa$ | electrolyte phase ionic conductivity |
| $\phi$ | electrical potential |
| $R$ | molar gas constant |
| $T$ | cell temperature |
| $F$ | Faraday constant |
| $i$ | current density |
| $I$ | external working current |
| $A$ | electrode plate area |
| $n$ | charge number of lithium ions |
| $\sigma$ | conductivity of solid phase |
| $i_0$ | exchange current density |
| $\alpha$ | transfer coefficients of reaction |
| $\eta$ | overpotential |
| $k_s$ | electrochemical reaction constant |
| $E_{\mathrm{OCV}}$ | electrode open circuit potential |
| $U$ | voltage of the cell |
| $R_{\mathrm{ext}}$ | extra ohm resistance |
| $R_{\mathrm{SEI}}$ | SEI film resistance |
| $c_{\mathrm{total}}$ | available lithium ions concentration |
| $CAP$ | maximum available capacity of the battery |
| $\bar{c}$ | average lithium ions concentration |
| $L$ | thickness of the micro cell |

*Subscripts*

| | |
|---|---|
| s | solid phase (active particles) |
| r | at the surface of the active particle |
| e | electrolyte phase |
| e-s | solid-electrolyte interface |
| max | maximum |
| t | terminal |
| p | positive electrode |
| sep | separator |
| n | negative electrode |
| a | anode |
| c | cathode |
| surf | the surface of the active particles |

*Superscripts*

| | |
|---|---|
| eff | effective |

*Abbreviations*

| | |
|---|---|
| LiBs | Lithium-ion batteries |
| EM | electrochemical model |
| ECM | equivalent circuit model |
| EVs | electric vehicles |
| SOH | state of health (for the battery) |
| EIS | electrochemical impedance spectroscopy |
| SOC | state of charge (for the battery) |
| PDEs | partial differential equations |
| EKF | extended kalman filter |
| P2D | pseudo-two-dimensional |
| HIL | hardware-in-the-loop |
| LS | least square |
| KF | kalman filter |
| GA | genetic algorithm |
| DST | dynamic stress test |
| FUDS | federal urban driving schedule) |
| GITT | galvanostatic intermittent titration technique |

identifies battery capacity, internal resistance and other parameters through the parameter identification method.

Nowadays the equivalent circuit model (ECM), electrochemical model (EM) and black box models are commonly used for estimation. Featured with the advantages of less computation and easy online realization, a large number of studies have already been done for SOH estimation based on the ECM. Xiong et al. [6] proposed a lumped parameter battery model to enhance the relationship of battery voltage to its state of charge (SOC) and capacity and used a multi scale extended kalman filter to get the battery capacity and SOC estimation. The results showed that the prediction accuracy was high and the computational efficiency had been greatly improved. The authors in [7,8] discussed the correlation between variation of parameters and degradation level and predicted the remaining capacity by selecting an appropriate fitting function with the fractional order impedance model. Although ECM has several merits for application, it has obvious shortcomings. It lacks physical meaning and ignores the internal microcosm reactions, thus, detailed information can't be depicted during battery aging process. The ECM has been studied to a certain degree, many research teams have contributed their effort to explore the battery management based on electrochemical model, which can be regarded as the state-of-the-art technique.

The EM is a kind of the first principle model which can not only reflect the changing process of the potential and the voltage, but also describe the reaction processes inside the battery. The model consists of serials of complicated, mutually coupled partial differential equations (PDEs) [9–12]. To improve computational efficiency and adjust the model to on-line application, the EM is usually simplified to some extent [13,14]. In [15], an adaptive output-injection observer based on the simplified EM was proposed to estimate the battery SOH, expressed in terms of the battery internal resistance. In addition to the capacity and internal resistance commonly used as SOH indicators, some electrochemical parameters can also represent the health state of the battery. The benefit of using these variables is that they can represent the health state of the battery independent of environmental conditions and use patterns. Extended kalman filter (EKF) [16], adaptive sliding mode observers (SMO) [17] and adaptive PDE observer [18] are used to estimate the number of cyclable Li-ions as an unknown battery parameter and clarified the superiority of regarding cyclable Li-ions as SOH indicator compared with other electrochemical variables. Solid phase diffusion time of Li-ions and some other parameters can also be used as SOH indicators. GK Prasad et al. [19] developed and simplified an EM that depended on two key aging parameters: cell resistance and positive solid phase diffusion time of Li-ions. The estimated parameters varied monotonically with aging, consistent with aging mechanism. The authors in [20] designed a multi-time-scale observer for SOC and SOH which is effective despite a range of common errors. Although a large amount of work had been done based on the EM, they didn't get the variation process of the chosen SOH indicators during the battery whole life-time. In other words, they lacked data support to depict the variation trend of the indicators.

The black box based model is another choice for SOH estimation.