



# A mechanism identification model based state-of-health diagnosis of lithium-ion batteries for energy storage applications

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## ABSTRACT

Advanced lithium-ion battery systems, in multi-cell configurations and larger-scale operations, are being currently developed for energy storage applications. Furthermore, the retired batteries are being increasingly second utilized in energy storage scenes. Thus, realistic and accurate battery state of health diagnosis and related aging mechanisms identification is desired to improve the battery management and control, and eventually guarantee the reliability and safety of the battery system. A half-cell model based battery state of health diagnostic method is proposed to investigate the aging mechanisms and possible attribute to the capacity fade in a quantitative manner. Using particle swarm optimization algorithm, the half-cell model is parameterized to quantify the battery degradation mechanisms derived from the parameter variations, which describe the electrode behavior with proper matching ratio and their evolutions at different battery aging levels. The reliability and robustness of the approach has been verified and evaluated by the database of the cells experienced different aging paths. Our approach is a data-model fusion method to offer the benefits of wide applicability to various cell chemistries and operating modes.

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## 1. Introduction

Battery systems are currently being designed in multi-cell configuration to fulfil the total voltage and power needs in diverse applications (Xiong et al., 2018a). Accordingly, there is a great possibility that the cells encounter aging path dependence problem owing to the adverse effect of cell inconsistency and complex, uncertain operation conditions (Xiong et al., 2018b; Ma et al., 2015). Unfortunately, battery performance is strongly relying on its aging state and previous degradation mechanism, which will eventually harm the reliability and security of the battery system. Furthermore, the batteries eliminated from EVs may be employed to the echelon-use application (Zhang et al., 2014). Thus, the accurate diagnosis of the battery aging path and associated degradation mechanisms is urgently needed to improve the battery prognostics and health management during operation, and to

provide a more detailed and realistic basis for battery screening and grouping in echelon-use application.

In order to quantify the health level of the battery, the most common indicator or notion in the literature is the state of health (SOH), which represents the specified performance and health state of a used battery compared to its fresh state (Rezvanianani et al., 2014). Despite the fact there is no clear definition of SOH, battery different features can be used to identify SOH, such as capacity and impedance, which correspond to the energy and power capability of the battery respectively (Waag et al., 2014). Afterwards the battery SOH diagnosis and estimation can be facilitated to the determination of the battery characteristic parameters (Rezvanianani et al., 2014; Waag et al., 2014).

Most of the methods reported in the literature, which are applied to evaluate the battery SOH indirectly or directly, are model based approaches combined with an optimization algorithm or filter to identify the parameter and state (Tao et al., 2017; Bercibar et al., 2016). In detailed, numerous optimization algorithms and filters have been included in the establishment of those model based approaches, such as extended Kalman filter (EKF) (Plett, 2004), sigma-point Kalman filter (SPKF) (Plett, 2006), Unscented

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Kalman filter (UKF) (Andre et al., 2013), dual EKF (Zou et al., 2015; Kim et al., 2012) or multi-scale EKF (Hu et al., 2012; Xiong et al., 2014), Genetic Algorithm (Chen et al., 2013), support vector machine (SVM) (Andre et al., 2013) and so on. In addition, the “black box” or data-driven based methods are also employed to evaluate the battery SOH. The authors in Ref. (Wei et al., 2011) present an estimation method based on Dempster-Shafer theory and the Bayesian Monte Carlo. A capacity estimation method utilizing improved sample entropy was proposed and evaluated by eight cells against aging and different temperatures (Hu et al., 2014). An advanced sparse Bayesian predictive modeling based SOH monitor is established considering temperature effects in Ref. (Hu et al., 2016). The authors in Ref. (Chaoui and Ibe-Ekeocha, 2017) presents an application of dynamically driven recurrent networks (DDRNs) for SOH estimation in online EV battery analysis. Nevertheless, the same change degree of capacity and impedance may differ in degradation mechanism inside the battery (Ma et al., 2015).

In order to be more explicit on such issues, many post-mortem analyses are carried out to reveal the battery degradation cause. However, they may lack potential when providing temporal resolution for battery diagnosis and protection during real operation applications, because these ex-suit methods involve the destruction and unrecoverable damage of the battery (Dubarry et al., 2012). To understand battery behavior and degradation by measuring only the voltage and current of the battery in different duty regimes, incremental capacity analysis (ICA) was advocated by Dubarry et al. (Dubarry et al., 2006, 2007, 2011; Dubarry and Liaw, 2009) recently to diagnose the battery aging state and related degradation mechanisms. This in-suit analysis technique has been verified for various Li-ion cell chemistries, including  $\text{Li}_x\text{Ni}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA) (Dubarry et al., 2006, 2007),  $\text{Li}_x\text{FePO}_4$  (LFP) (Dubarry and Liaw, 2009),  $\text{Li}_x\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$  and  $\text{Li}_x\text{Mn}_2\text{O}_4$  composite (NMC + LMO) (Dubarry et al., 2011). Differential voltage analysis (DVA), which also belongs to in-suit technique, was used by Honkura et al. (2011) and Bloom et al., 2005, 2006, 2010 to derive time related aging behavior in a quantitative manner. ICA and DVA are used to analyze the battery aging path dependence quantitatively specific to Beijing pure electric bus in our previous work (Ma et al., 2015).

However, it is hard to figure out the contribution of positive and negative electrode behavior to the cell degradation separately, because the variations of the features in the IC and DV curves may come from both electrodes in the cell. Dubarry et al. (2012) proposed a half-cell data based model to understand the evolution of the IC and DV curves and simulate a variety of “what if” situations for the quantitative analysis of the electrode contributions to different cell aging modes. Referring to the concept of this approach, we propose a half-cell model to simulate and track the cell aging behavior by optimal calculation, in which the outputs of the simulation can be used to investigate cell degradation mechanisms quantitatively by the combination of the electrode behavior and full cell voltage curve. This model is not only a learning tool to help us understand how battery degrades, but also an effective diagnostic approach to reveal battery aging mechanisms in real applications.

### 1.1. Contribution of the paper

A key contribution of this study is that a battery SOH diagnosis approach based on open-circuit-voltage analysis and half-cell model fusion was developed, thus battery degradation mechanisms and their contribution to the battery capacity fade were investigated in a quantitative manner. In our approach, the PSO algorithm is employed to identify the half-cell model parameters

based on the electrode behavior and battery performance at different aging levels, which are applied to diagnose the battery degradation with the fusion of the OCV analysis. In addition, the ICA and DVA combined method is applied to analyze the OCV variations. Our approach offers a unique simulation capability with mechanistic understanding of battery fade to address aging path dependence, which offers the benefits of wide applicability to various cell chemistries.

### 1.2. Organization of the paper

The remainder of the paper is organized as follows: Section 2 describes the half-cell model and model parameter identification method based on PSO. And battery SOH diagnosis approach based on OCV analysis and half-cell model fusion is also presented in this Section. To evaluate the proposed approach, eight commercial lithium-ion cells are used to establish the cycle life test database in the study. The experiment and results are described in Section 3 and 4. The evaluation of the proposed approach is discussed in Section 5 before conclusions are drawn in Section 6.

## 2. Synthesized battery state-of-health diagnostic approach

### 2.1. Half-cell model descriptions

The half-cell model developed in the paper consists of two layers: a bottom layer using half-cell data that characterize the positive and negative electrode behavior and a top layer that describe cell configuration and performance at different aging stages. The bottom layer employs two separated half-cell data to handle the electrode behavior against voltage and capacity variation. The top layer builds the relationship between two electrodes and a full cell by using equations, considering electrode composition, material properties and ratio relation as a function of degradation level. The half-cell model is configured to simulate and track the cell aging behavior by optimal calculation, in which the cell performances in aging are also inputs to the simulation. The outputs of the simulation, in other words the model parameters, are capacity and initial SOC variations of two electrodes as a function of aging that reflect the changes of electrode composition and their ratio relation. Then cell degradation modes and mechanisms can be determined in a quantitative manner.

The architecture of the half-cell model in this work is shown in Fig. 1. Our model accommodates electrode-specific phenomena, in which the cell and its internal electrode behavior are combined and applied to the model calculation. And the model outputs can directly quantify the cell degradation mechanisms at different aging stages. The model is built in MATLAB to simulate cell degradation mechanisms with architecture in the configuration of effective computation, layer integration, and flexibility.

### 2.2. Model parameter identification based on particle swarm optimization

The PSO algorithm starts with a random given initial value, and obtains a different solution by a certain number of iterations. Finally, the optimal solution is obtained by using the fitness value as the evaluation index. In  $m$  dimensional search space, the population  $\mathbf{X}=(X_1, X_2, \dots, X_i, \dots, X_n)^T$  is composed of  $n$  particles, in which the velocity and individual extremum of the particle  $i$  are  $\mathbf{V}_i=(V_{i1}, V_{i2}, \dots, V_{id}, \dots, V_{im})^T$  and  $\mathbf{P}_i=(P_{i1}, P_{i2}, \dots, P_{id}, \dots, P_{im})^T$  respectively, and global extremum is  $\mathbf{P}_g=(P_{g1}, P_{g2}, \dots, P_{gd}, \dots, P_{gm})^T$ . In each iteration, the velocity and position of the particle  $i$  are updated by Eq. (1) and Eq. (2).

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