A Sensitivity-Based Group-Wise Parameter Identification Algorithm for the Electric Model of Li-Ion Battery

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ABSTRACT
This paper proposes a sensitivity-based group-wise parameter identification algorithm for the electrical model of Li-ion battery. A global sensitivity analysis method is first performed in the entire parameter space to evaluate the identifiability of the model parameters. Then, the parameters are sorted and grouped by the global sensitivity indices. Finally, a group-wise method embedded with the Levenberg–Marquardt algorithm is followed to identify the parameters. Numerical simulation results and comparisons demonstrate that the proposed group-wise identification algorithm can serve as a reliable tool for extracting parameters.

INDEX TERMS
Li-ion battery, sensitivity analysis, parameter identification, group-wise identification, Levenberg-Marquardt algorithm.

I. INTRODUCTION
Increasing concerns for the environment have promoted the popularization of electrical vehicles (EVs) and hybrid electrical vehicles (HEVs). LIBs are especially suitable for powering EVs and HEVs given their high-energy density and environmental friendliness [1]–[3]. In the past few years, much attention has been paid to the modeling and simulation of LIBs from a systems engineering point of view [1].

Elaborate models have been built to predict the electric performance of LIB: data-based model [4]–[6], equivalent circuit model [7]–[9], electric-distribution model (e.g., two-dimensional potential and current density distribution (PDD) model [10], [11]) and electro-chemical model (e.g., single particle (SP) model [12], pseudo-two-dimensional (P2D) model [13], [14], and multi-scale multi-dimensional (MSMD) model [15]). Among various battery models, the PDD model and MSMD model are better at describing the distributed electrical and thermal behavior, as the electrical and thermal behavior is quite non-uniform through the cells during the charge/discharge operations at high current rates [16]. MSMD combines the electrochemical-, electrical-, and thermal-coupled physics in multi-domains and is considered as a rigorous model to describe the battery behavior [15]. While PDD is relative simpler than MSMD by considering only Ohm’s law and charge conservation on the electrodes based on the simplified polarization characteristics of the electrodes [10], [11]. This feature has made the PDD model widely used in the industry [15], [17].

The accuracy of parameters plays a critical role in the model based prediction of the battery performance. Many algorithms including recursive least squares method [18], non-recursive least squares method [19], Kalman filter method [20], [21], convex optimization method [22] and evolutionary algorithm [23], [24] have been proposed for the parameter estimation or identification of LIBs. Among these methods, the recursive least squares methods and the evolutionary algorithms have been applied to identify the parameters for the electric-distribution model and electrochemical model. For example, the Levenberg-Marquardt (LM) algorithm is employed to identify the parameters of the SPM [18]. GA [23] is utilized to solve the parameter identification of the P2D model, and the PSO algorithm is also applied to identify the parameters of the electrochemical model [24]. However, these works identify all the parameters synchronously without considering the importance of parameters. This may lead to a situation that important parameters cannot deserve
enough attention, while unimportant parameters consume computing resources excessively.

On the other hand, Ramadesigan et al. [1] have pointed out that general numerical algorithms for optimization can get stuck in local optima due to a large number of parameters and a very large searching space. To address the stagnation of local optima, a concept of sequential step-by-step identification approach is proposed in [1] to identify the model parameters for the maximization of the energy density of LIB. It has also been pointed out that the suggested approach will tend to have better convergence if the parameters are ranked by their importance. One method of evaluating the parameter importance is sensitivity analysis (SA), which investigates the effect of parameter change in the output of mathematical models. A sensitivity based stepwise parameter identification approach is proposed in [25] and [26]. A local sensitivity analysis (LSA) method is combined to classify the parameters into strongly sensitive, conditional sensitive and weakly sensitive parameters. However, LSA explores only a small fraction of the parameter space by studying the impact of small input perturbation on the model output and is not suit for high dimensional and nonlinear system. To overcome the limitations (linearity and local variations) of LSA, a global sensitivity analysis (GSA) method is proposed to utilize a global set of samples to explore the parameter space. GSA is applied to define the parameter importance for fuel cell models [27]. Up to now, the GSA-based importance analysis has not been proposed for the LIB models yet. The GSA methods include Elementary Effects Test (EET), Regional Sensitivity Analysis (RSA), Fourier Amplitude Sensitivity Test (FAST), and Variance Based Sensitivity Analysis (VBSA) [28]. Among these methods, VBSA has been assessed as versatile and effective method for sensitivity analysis of complex nonlinear models [29]–[31], as it takes factor correlations into account and explores their model over different combinations of values for the uncertain inputs. As the two-dimensional uneven PDD model is a time/space coupled distributed parameter system (DPS) with very high nonlinearity and high correlation [32], VBSA is employed here to analyze the parameter sensitivities for this model.

Up to now, few works have been proposed for the parameter identification of the PDD model. The “art” of trial and error is adopted to determine the value of model parameters [11], which requires exhaustive enumeration. In this paper, a sensitivity-based group-wise parameter identification algorithm is proposed to identify the parameters of the PDD model asynchronously. VBSA [29] is first adopted to calculate the global sensitivity indices of the parameters of the PDD model. Based on the dimensionless sensitivity indices, all the parameters are sorted in descending order. The parameters with similar sensitivities are grouped together, and then identified group by group with the Levenberg-Marquardt (LM) algorithm. As LM is a statistical algorithm with gradient-based nonlinear least squares regression, and can converge quickly to the optima [33]–[35].

The rest of this paper is organized as follows: Section II introduces the PDD model and the synchronous parameter identification complexity. Section III proposes a sensitivity-based group-wise identification algorithm to address this complexity. The sensitivity indices of the parameters and simulation results with the proposed identification approach are presented in Section IV. Finally, Section V offers some concluding remarks.

II. MATHEMATICAL MODEL FOR LITHIUM-ION BATTERY AND PROBLEM FORMULATION

A simple cell that consists of two parallel plate electrodes with very small distance is considered in this paper. Fig. 1 shows a schematic diagram of the current flow in the cell during discharge. As the distance between electrodes is extremely small, the current flow between electrodes is assumed to be perpendicular to the electrodes (see Fig. 1). The potential distributions in positive and negative electrodes are governed by the following two-dimensional PDD model (see [36]):

\[
\nabla^2 \phi_p(\psi, t) = -r_p J_p(\psi, t) \quad \text{in } \Omega_p \\
\nabla^2 \phi_n(\psi, t) = +r_n J_n(\psi, t) \quad \text{in } \Omega_n
\]

(1)

(2)

where the Laplace operator \(\nabla^2 = (\partial^2/\partial y^2 + \partial^2/\partial z^2)\), \(\phi_p(\psi, t)\) and \(\phi_n(\psi, t)\) are potentials (V) of the positive and negative electrodes, respectively, \(r_p\) and \(r_n\) are resistances (Ω) of the positive and negative electrodes, respectively, \(J_p(\psi, t)\) is the current density, which is current per unit area (A m\(^{-2}\)). \(\Omega_p\) and \(\Omega_n\) are domains of the positive and negative electrodes, respectively. For the relevant boundary conditions of \(\phi_p(\psi, t)\) and \(\phi_n(\psi, t)\), please refer to [36].

The current density \(J_c(\psi, t)\) can be written as a polarization expression of the potential difference between positive and negative electrodes and expressed as [37] and [38].

\[
J_c(\psi, t) = Y_c(\psi, t)(\phi_p(\psi, t) - \phi_n(\psi, t) - U(\psi, t))
\]

(3)
where $\phi_p(\psi, t) - \phi_c(\psi, t)$ is the potential difference. In the experiment of determining the polarization characteristic for the battery by Gu [39], $U(\psi, t)$ is the intercept of the voltage-current curve, unit (V); $Y_c(\psi, t)$ is the inverse of the slope of the voltage-current curve, unit ($S/cm^2$). Gu declared that according to the experimental data, both $U(\psi, t)$ and $Y_c(\psi, t)$ varied with respect to the depth of discharge (DOD), the dependencies can be expressed as a function of DOD:

$$U(\psi, t) = x_1 + x_2D(\psi, t) + x_3D(\psi, t)^2 + x_4D(\psi, t)^3$$

(4)

$$Y_c(\psi, t) = x_5 + x_6D(\psi, t) + x_7D(\psi, t)^2$$

(5)

where $x_1, x_2, \ldots, x_7$ are the fitting parameters and $D(\psi, t)$ is the distribution of DOD on the electrodes, which is calculated from the integration of $J_c(\psi, t)$ as [10], [11], [36]:

$$D(\psi, t) = \frac{1}{\tau} \int_{t_0}^{t} J_c(\psi, t)dt$$

(6)

where $t$ is the discharge time (s), $t_0$ is the discharge starting time (s), and $\tau$ is the theoretical capacity per unit area ($\text{Ah m}^{-2}$) of the electrodes. Set $x \triangleq [x_1, x_2, \ldots, x_7]^T$ for brevity.

Much attention has been given in the parameter identification of LIB models, leading to many approaches for effective identification methods. However, the identified algorithms in many of these works identify all parameters synchronously. Due to the large number of parameters and the very large searching space, the optimal solution to the parameter identification problem of LIB model will be difficult to obtain. On the other hand, it is computationally expensive to identify all parameters simultaneously because of the interdependence of parameters. To overcome the drawback of synchronous identification algorithms, a feasible approach is to develop identification algorithms asynchronously identifying the parameters. For the asynchronous identification, three follow-up questions are required to be addressed:

1) The identification order of parameters;
2) The parameter size for each step of identification;
3) The switch among parameter identification processes.

The main objective of this paper is to develop an asynchronous parameter identification algorithm to determine the value of the parameter $x$ in the battery electric model (1)-(6). In the proposed identification algorithm, the three mentioned questions will be well addressed.

### III. SENSITIVITY-BASED GROUP-WISE PARAMETER IDENTIFICATION

This section provides the sensitivity-based group-wise parameter identification algorithm for the electric model (1)-(6), where the block diagram of the suggested identification algorithm is illustrated in Fig. 2. In the proposed identification algorithm, three questions of asynchronous identification given in Section II are solved as follows, respectively.

1) The sensitivity analysis of the parameters is employed to determine the identification order;
2) The parameters of similar sensitivities are grouped together and then identified synchronously by using the LM algorithm;
3) The parameter identification procedures are switched between groups according to the value of the objective/fitness function given in later.

#### A. SENSITIVITY ANALYSIS

Parameter identifiability can be indicated by its sensitivity [25]. The sensitivity quantifies the importance of individual parameter by evaluating its contribution to the model output. As shown in Fig. 3(a), the length of principal axis illustrates the sensitivity indices of parameters in the hyperspace, that is, the strongest sensitive parameter is indexed by the longest principal axis in the hyperspace. To intuitively illustrate the sensitivity-based parameter importance analysis, the parameters are projected to a two-dimensional plane and resorted according to the value of their sensitivity indices (see Fig. 3(b)).

In this paper, a global sensitivity analysis method, VBSA [28], [29], is used to analyze the sensitivity of the seven parameters in the battery model (1)-(6). According to the sensitivity analysis result of these seven parameters, they are processed as follows:

1) Seven parameters are sorted from large to small by the value of their sensitivity indices.
2) The parameters of similar sensitivity indices are grouped together.

The main procedure of VBSA for this model is presented in the Appendix.
B. GROUP-WISE IDENTIFICATION PROCEDURE

As the strongly-sensitive parameters dominate the model outputs, the identification should be conducted from the strongly sensitive groups to weakly sensitive groups. The group-wise identification procedure embedded with a switching strategy of parameter groups is presented as follows:

1) The identification is started from the first group of parameters with the LM algorithm.

2) A weighted objective/fitness function $f$ in (7) is calculated after each iteration, which is a criterion for switching between different parameter groups.

3) For the $g$-th parameter group identification, once the difference of the objective function value between two iterations $\tau_g$ (equals to $f^g_{\phi_{g}} - f^{g+1}_{\phi_{g}}$) is less than the preset value $\tau_{set}$, the parameter of the $g$-th group is fixed with the identified value.

4) The identification procedure then switches to the $(g + 1)$-th parameter group.

Weighted objective function: the weighted objective/fitness function $f$ (i.e., the performance evaluator) is constructed for the identification of the parameter vector $x$ in the battery electric model (1)-(6), which is a quadratic function of the difference between the experimental terminal voltage $\hat{\phi}_k$ and the predicted voltage $\phi_k$ with additional parameters $w_k > 0$, $k \in \{1, 2, \cdots, N\}$:

$$f(\phi_k) = \sum_{k=1}^{N} w_k (\phi_k - \hat{\phi}_k)^2 = (\phi - \hat{\phi})^T W (\phi - \hat{\phi})$$ (7)

where $N$ is the number of fitting curves, $\phi \triangleq [\phi_1, \phi_2, \cdots, \phi_N]^T$ and $\hat{\phi}$ is the estimation of $\phi$, both are vectors consisting of $\phi_k$ and $\hat{\phi}_k$, $k \in \{1, 2, \cdots, N\}$, respectively. $\hat{\phi}_k$ is predicted terminal voltage obtained by calculating the difference between $\hat{\phi}_k$ and $\phi_k$ in the tab. $W \triangleq \text{diag}(w_1, w_2, \cdots, w_N)$ is the weight matrix assigned to obtain a better fitness.

In what follows, we will define the specified form of the weight matrix $W$, whose element $w_k$ is applied to the $k$-th experimental terminal voltage curve and chosen as:

$$w_k = \gamma_k \chi_k$$ (8)

where $\gamma_k$ takes into account the experimental deviations in the $k$-th voltage curve and is defined as

$$\gamma_k = \frac{1}{\frac{1}{\sigma_k^2} \sum_{i=1}^{N} \sum_{j=1}^{N_k} \eta_j}$$ (9)

where $N_k$ is the number of experimental data points in the $k$-th voltage curve, $\sigma_k \triangleq \sqrt{\frac{1}{N_k-1} \sum_{j=1}^{N_k} (\hat{\phi}_{k,j} - \phi_{k,j})^2}$, in which $\hat{\phi}_{k,j}$ is the predicted voltage of the $j$-th point in the $k$-th curve, $\phi_{k,j}$ is the experimental voltage at the $j$-th point in the $k$-th curve. The parameter $\gamma_k$ is proportional to the difference between the previous voltage predictions and the experimental data in the $k$-th voltage curve, shown as

$$\chi_k = 1 + \frac{(w_k, \max - 1) \eta_k}{\eta_k, \max}$$ (10)

where $w_k, \max > 1$ is a hyper parameter which is assigned arbitrarily in [18]. $\eta_k \| \hat{\phi}_k - \phi_k \|$, $\eta_k, \max$ is the maximum value of $\eta_k$ for each $k \in \{1, 2, \cdots, N\}$. Obviously, the parameter $\chi_k$ varies between 1 and $w_k, \max$, that is, $\chi_k = 1$ for the best fitting point and $\chi_k = w_k, \max$ for the worst fitting one.

Algorithm 1 Sensitivity-Based Group-Wise Identification

1. Initialize parameter vector $x$, optimization range, generate sensitivity analysis samples, objective value $f_{set}$;
2. Calculate the sensitivity index $S_{\phi_{k}}$ and $S_{\lambda}$ by VBSA method;
3. The parameters of similar sensitivities are grouped together (e.g. $G$ groups);
4. repeat
5. for $g = 1, G$ do
6. Optimize the $g$-th group parameter by LM, evaluate the difference $\tau_g$ between two iterations;
7. if $\tau_g \leq \tau_{set}$ then
8. Set the $g$-th group parameter to the optimized value, Evaluate the terminal voltage $\hat{\phi}_k$ and objective function $f_g (\phi)$ according to (7);
9. end if
10. end for
11. until the stop criterion $f_g \leq f_{set}$ is satisfied.

C. LM ALGORITHM

The LM algorithm [40] is applied to solve nonlinear least squares problems, and is embedded in the procedure to identify each group of parameters. LM can adaptively adjust itself to the gradient-descent method when the parameters are far away from the optimal values, and adjust itself to the Gauss-Newton method when the parameters are approaching the optimal values. LM updates the parameters by a correction vector $\Delta x$ according to the objective function $f$ (see (7)).

$$\Delta x = (J^T W J + \lambda I)^{-1} J^T W (\phi - \hat{\phi})$$ (11)

where $J$ is the matrix of partial derivatives of the terminal voltage with respect to the fitting parameter vector $x$ evaluated at all the experimental voltage data. LM adaptively alters the algorithmic parameter value $\lambda$ updates between the gradient descent one and the Gauss-Newton one. The parameter $\lambda$ determines how the LM algorithm works and is initialized to be large. If the iteration results in a better approximation, then the parameter $\lambda$ is decreased to 0.1$\lambda$, and LM is more like a Gauss-Newton update one. If the iteration provides a worse approximation, then the parameter $\lambda$ is increased to 10$\lambda$, and LM approaches that of the gradient descent update. The details of LM can refer to [33].
The detailed framework of the suggested sensitivity-based group-wise algorithm is shown in Algorithm 1. The group-wise identification procedure can be found from step 5 to step 10.

IV. EXPERIMENTAL VALIDATION

A. EXPERIMENTAL SETUP

A lithium-ion battery (LiFePO4 prismatic cell) was used in the discharge tests at 25 ± 0.2°C in a thermal chamber. The nameplate capacity of the LIB is 60Ah. The discharge tests were done using a battery testing system (BTS-300A/60V). The experimental platform is shown in Fig. 4(a). The schematic diagram of the signal and data flow for the experimental platform is also presented in Fig. 4(a). Fig. 4(b) shows the dimensions of electrodes and positions of the tabs of the prismatic battery. The cell is discharged at various rates (1C (60A)), 2C (120A), 3C (180A)) from its fully charged state to fully discharged state. The experimental terminal voltage data is collected per second during its discharge.

B. NUMERICAL CALCULATION AND PARAMETER SETUP

The numerical solutions of the battery electric model (1)-(6) subject to the associated boundary conditions are obtained by COMSOL, which is a commercial software package for accurate numerical simulation of partial differential equations (PDEs) using the finite element method (FEM). The simulation results and identified parameters are exchanged through the interface of COMSOL with MATLAB. In the proposed group-wise algorithm, set \( \lambda = 0.1 \), \( \tau_{set} = 0.001 \), \( N = 3 \) and for the weight matrix \( W \), set, \( w_{1,max} = 4 \), \( w_{2,max} = 8 \), and \( w_{3,max} = 10 \).

C. SENSITIVITY-BASED PARAMETER IMPORTANCE INDEX

The main-effect index \( S_{i}^{M} \) and the total-effect index \( S_{i}^{T} \) of sensitivity for parameter \( x \) are calculated by (A.4) and (A.7), respectively. The values of the sensitivity indices are shown in Table 1. Fig. 5 is a graphic illustration of Table 1. As shown in Fig. 5, the values of the sensitivity indices of parameter \( x_4 \) and \( x_3 \) are larger than those of other parameters. This means they are more important than other parameters for the output of the model. The sum of main-effect indices is 0.7899, while the sum of total-effect indices is 2.0839. These two sums are not equal to 1, which is caused by the interactions of the parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>( S_{i}^{M} )</th>
<th>( S_{i}^{T} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( x_1 )</td>
<td>0.0357</td>
<td>0.1677</td>
</tr>
<tr>
<td>( x_2 )</td>
<td>0.0543</td>
<td>0.3954</td>
</tr>
<tr>
<td>( x_3 )</td>
<td>0.3063</td>
<td>0.5115</td>
</tr>
<tr>
<td>( x_4 )</td>
<td>0.2691</td>
<td>0.5235</td>
</tr>
<tr>
<td>( x_5 )</td>
<td>0.0439</td>
<td>0.1595</td>
</tr>
<tr>
<td>( x_6 )</td>
<td>0.0352</td>
<td>0.1685</td>
</tr>
<tr>
<td>( x_7 )</td>
<td>0.0454</td>
<td>0.1578</td>
</tr>
<tr>
<td>( \sum )</td>
<td>0.7899</td>
<td>2.0839</td>
</tr>
</tbody>
</table>

The sensitivity indices stated in Fig. 5 is calculated by an initial sample size of \( m = 1000 \) for parameter vector \( x \). Fig. 6 gives the evolution of the total sensitivity index \( S_{i}^{T} \) with the increasing of sample size. This verifies the effectiveness of the parameter classification in Table 2.

Some negative indices are found in Fig. 7, which are caused by a function of the Monte Carlo “short-cut” examination.
adopted in VBSA [41]. The sample size in the horizontal axis is a function of the initial sample size \(m\) and the length of parameter vector \(x\), which is equivalent to 9000 (= 1000 \times (7 + 2)).

The parameters are then classified into 3 groups by their values of sensitivity indices (see Table 2). For the convenience of comparison, three groups of parameter values are normalized to illustrate the variation of parameters during identification (Fig. 7). It is easier to identify the strongly sensitive parameters (Fig. 7(a)) than the weakly sensitive ones (Fig. 7(c)) with fewer iterations. As the weakly sensitive parameter has very little effect on the model outputs, which requires more iterations before finding the optimal value.

The value of the parameter vector \(x\) in the battery electric model (1)-(6) is identified by the suggested group-wise identification algorithm through fitting the three experimental terminal voltage curves simultaneously. Table 3 gives the identified value of the parameter vector \(x\). As shown in Fig. 8(a), the predictions with identified parameter vector \(x\) and the measurements of the terminal voltage curves are well matched each other, and this demonstrates the effectiveness of the proposed algorithm. Fig. 8(b) depicts the corresponding trajectory of the weight matrix \(W = \text{diag}(w_1, w_2, w_3)\).
The potential distributions on the positive and negative electrodes during discharge are obtained as a function of time for various discharge rates of 1C, 2C and 3C. Fig. 8(a) shows the potential distributions on the positive electrode with discharge rate of 2C at discharging time $t = 20$ min. Since all the current flows into the tab from the entire electrode plate, the potential gradient on the positive electrode shown in Fig. 9(a) is seen to be most severe in the region near the tab. Fig. 9(b) indicates the potential distribution on the negative electrode, where the potential gradient is the strongest at the region near
FIGURE 10. The distributions of (a) current density $J_c$ (A/m$^2$) and (b) DOD at the discharging time of 20min during discharge at a rate of $2C$.

TABLE 3. The identified value and 95% confidence interval of the parameter vector $x$.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Identified value</th>
<th>95% Confidence interval</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x_1$</td>
<td>3.210</td>
<td>$3.210 \pm 2.701 \times 10^{-3}$</td>
</tr>
<tr>
<td>$x_2$</td>
<td>$-3.235 \times 10^4$</td>
<td>$(-3.235 \pm 0.179) \times 10^4$</td>
</tr>
<tr>
<td>$x_3$</td>
<td>1.693</td>
<td>$1.093 \pm 6.1 \times 10^{-2}$</td>
</tr>
<tr>
<td>$x_4$</td>
<td>$-1.680$</td>
<td>$-1.680 \pm 5.31 \times 10^{-3}$</td>
</tr>
<tr>
<td>$x_5$</td>
<td>$1.370 \times 10^3$</td>
<td>$1.370 \pm 1.083$</td>
</tr>
<tr>
<td>$x_6$</td>
<td>$6.908 \times 10^3$</td>
<td>$(-6.908 \pm 0.879) \times 10^3$</td>
</tr>
<tr>
<td>$x_7$</td>
<td>$-1.630 \times 10^2$</td>
<td>$(-1.630 \pm 0.134) \times 10^2$</td>
</tr>
</tbody>
</table>

Tab, since all the current should flow from the tab through the entire electrode plate. Fig. 10(a) depicts the current density $J_c$ flows from the negative electrode to the positive electrode with a discharge rate of $2C$ at discharging time of 20min, while Fig. 10(b) illustrates the uneven distribution of DOD.

To demonstrate the advantage of the proposed group-wise algorithm, a comparison study is also conducted among the method of trial and error depicted in [11], group-wise algorithm with $W = \text{diag}\{1, 1, 1\}$, and weighted group-wise algorithm. Fig. 8 also compares the experimental and predicted terminal voltage curves at discharge rates of $1C$, $2C$, and $3C$ through the multi-curve fit by group-wise algorithm with $W = \text{diag}\{1, 1, 1\}$ (Fig. 8(c)) and method of trial and error (see Fig. 8(d)). It is clear that the proposed weighted sensitivity based group-wise algorithm provides more accurate discharging curve fitness compared to group-wise algorithm with $W = \text{diag}\{1, 1, 1\}$ and method of trial and error. Moreover, although the fitting result of group-wise algorithm with $W = \text{diag}\{1, 1, 1\}$ is better than that of trial and error, it is worse than that of weighted group-wise algorithm. This is because the same weights assigned to different curves but without considering the difference of the data number of each voltage curve (e.g., there are 2860 data points for the $1C$-discharging voltage curve, 1360 data points for $2C$ and 850 for $3C$).

95% confidence interval: From a statistical point of view, it is very useful to obtain the confidence interval instead of point estimation for the fitting parameter vector $x$. In this paper, the 95% confidence interval of the parameter vector $x$ is calculated as follows

$$x_i^* - T(1-0.05/2)SE \sqrt{a_{ii}} \leq x_i \leq x_i^* + T(1-0.05/2)SE \sqrt{a_{ii}}$$

(12)

where $x_i^*$ is the point estimation of the parameter $x_i$, $i \in \{1, 2, \ldots, 7\}$, $T(1-0.05/2)$ is a value of $T$-distribution with $(N - 7)$ degrees of freedom, $SE$ is the standard deviation and is calculated by

$$SE = \sqrt{\frac{1}{N-7} \sum_{\varsigma=1}^{N} \sum_{j=1}^{N_{\varsigma}} (\phi_{\varsigma,j} - \hat{\phi}_{\varsigma,j})^2}$$

$N$ is the number of experimental data points and derived by

$$N = \sum_{\varsigma=1}^{N} N_{\varsigma},$$

and $a_{ii}$ is the $i$-th main diagonal element of the matrix $(J^TJ)^{-1}$. The 95% joint confidence intervals of all seven parameters obtained from the simultaneous fit are given in Table 3. In order to illustrate the goodness of the voltage curve predictions by using the value of the parameter vector $x$ defined by the confidence interval, Fig. 11 provides a graphic illustration of the 95% confidence interval of the parameter $x_1$. It can be observed from Table 3 and Fig. 11 that the confidence interval is appropriate, and it is declared that the good confidence intervals for the parameter values are indicative of the fact that the estimation does yield one set of parameters that are significant [18]. The narrower confidence interval indicates this identification study has reasonable
FIGURE 11. Graphic illustration of the 95% confidence interval of the parameter \( x_1 \) at different discharge rates (a) 1C, (b) 2C, (c) 3C. Point estimates obtained from the identification are used for the rest parameters. LCIL and UCIL represent the lower and upper confidence interval limits, respectively.

V. CONCLUSION
Synchronous identification for the nonlinear spatiotemporal coupled system is complicated because of the interdependence of parameters. The sensitivity-based group-wise parameter identification algorithm has addressed this complexity. The VBSA method measures the global sensitivity indices of each parameter. Parameters with similar sensitivities are grouped together. The group-wise method embedded with the LM algorithm is applied to identify the parameters. The proposed identification approach can efficiently capture the dominant characteristic of the system output with relatively small number of model evaluations, and overcome the drawback of the local minimum entrapment of the synchronous parameter identification. The experimental result demonstrates the effectiveness of this approach.

The proposed sensitivity-based group-wise method combined with PDD model can be applied to modify the configuration of electrodes (e.g., the dimension of the electrodes, the location of tab, etc.) to assure the uniformity of potential distribution and temperature distribution if coupled with a thermal model. The proposed framework is also suited to analyze and identify the physical parameters for the rigorous electrochemical models such as MSMD model. Moreover, for the nonlinear distributed parameter systems with large amount of parameters, the proposed identification method can be easily implemented to address the problems of over-parameterization and over-fitting.

APPENDIX
The main procedure of VBSA is summarized as follows:

Parameter Sampling: Latin hypercube sampling (LHS) [43] is applied to initialize parameters by dividing the parameter space of \( x_i \) \( (i = 1, \ldots, 7) \) into \( m \) classes of equal probability, two independent parameter matrices \( H \) and \( P \) are generated in (A.1).

\[
H = \begin{bmatrix}
  h_{11} & h_{12} & \cdots & h_{1n} \\
  h_{21} & h_{22} & \cdots & h_{2n} \\
  \vdots & \vdots & \ddots & \vdots \\
  h_{m1} & h_{m2} & \cdots & h_{mn}
\end{bmatrix}
\]

\[
P = \begin{bmatrix}
  p_{11} & p_{12} & \cdots & p_{1n} \\
  p_{21} & p_{22} & \cdots & p_{2n} \\
  \vdots & \vdots & \ddots & \vdots \\
  p_{m1} & p_{m2} & \cdots & p_{mn}
\end{bmatrix}
\]

where \( m = 1000 \), \( n = 7 \)

New Matrix Construction: \( H_p^{(i)} \) is obtained by replacing the \( i \)-th column of matrix \( H \) with the \( i \)-th column of matrix \( P \). \( P_H^{(i)} \) is obtained by replacing the \( i \)-th column of matrix \( P \) with the \( i \)-th column of matrix \( H \).

\[
H_p^{(i)} = \begin{bmatrix}
  h_{11} & h_{12} & \cdots & p_{1i} & \cdots & h_{1n} \\
  h_{21} & h_{22} & \cdots & p_{2i} & \cdots & h_{2n} \\
  \vdots & \vdots & \ddots & \vdots & \ddots & \vdots \\
  h_{m1} & h_{m2} & \cdots & p_{mi} & \cdots & h_{mn}
\end{bmatrix}
\]

\[
P_H^{(i)} = \begin{bmatrix}
  p_{11} & p_{12} & \cdots & h_{1i} & \cdots & p_{1n} \\
  p_{21} & p_{22} & \cdots & h_{2i} & \cdots & p_{2n} \\
  \vdots & \vdots & \ddots & \vdots & \ddots & \vdots \\
  p_{m1} & p_{m2} & \cdots & h_{mi} & \cdots & p_{mn}
\end{bmatrix}
\]

Model Evaluation: The model outputs \( \hat{\phi}(H) \), \( \hat{\phi}(P) \), \( \hat{\phi}(H_p^{(i)}) \) and \( \hat{\phi}(P_H^{(i)}) \) are evaluated for the parameter matrices in (A.1) and (A.2). These outputs need to be further transformed into a scalar for the sensitivity analysis.

certainty. While the estimates with wide confidence interval is not precise [42].
The dimensionless Nash and Sutcliffe efficiency (NSE) is adopted for this transformation [44].

\[ f(x) = 1 - \frac{\sum_i (\phi - \hat{\phi}(x))^2}{\sum_i (\phi - \phi)^2} \quad (A.3) \]

where \( \phi \) is the experimental terminal voltage, \( \hat{\phi} \) is the mean of the experimental voltage, and \( \phi(x) \) is the predicted voltage with sampled parameter vector \( x \). The NSE coefficient ranges from \(-\infty \) to 1. An efficiency of 1 (\( f = 1 \)) corresponds to a perfect match of the modeled voltage \( \phi(x) \) to the measured voltage \( \phi \).

**Sensitivity calculation:** there are two indices to measure the sensitivity: the main-effect index \( S_M^f \) and the total-effect index \( S_T^f \). \( S_M^f \) measures the contribution of \( x_i \) to the model output by varying \( x_i \) alone. It is presented as follows:

\[ S_M^f = \frac{V_{x_i}(E_{X\sim\{i\}}[f|x_i])}{V(f)} \quad (A.4) \]

where \( X\sim\{i\} \) is the matrix of all parameters but \( x_i \), and \( V_{x_i}(E_{X\sim\{i\}}[f|x_i]) \) is the expected reduction in variance that would be obtained if \( x_i \) could be fixed, and is obtained from

\[ V_{x_i}(E_{X\sim\{i\}}[f|x_i]) = \frac{1}{N} \sum_{j=1}^{N} f(H_j)f(P_j^{(i)})-f_0^2 \quad (A.5) \]

and in which \( f_0 = \frac{1}{N} \sum_{j=1}^{N} f(H_j) \), \( H_j \) is a parameter vector which denotes the \( j \)-th row of matrix \( H \). \( V(f) \) in (A.4) is calculated as

\[ V(f) = \frac{1}{N} \sum_{j=1}^{N} (f(H_j))^2 - \left( \frac{1}{N} \sum_{j=1}^{N} f(H_j) \right)^2 \quad (A.6) \]

\( S_T^f \) measures the contribution of \( x_i \) to the output by including all the variance attributed to its interactions with other parameters. It is given by

\[ S_T^f = 1 - \frac{V_{X\sim\{i\}}(E_{x_i}[f|X\sim\{i\}])}{V(f)} \quad (A.7) \]

where \( V_{X\sim\{i\}}(E_{x_i}[f|X\sim\{i\}]) \) is the expected reduction in variance that would be obtained if all parameters except \( x_i \) could be fixed and defined by

\[ V_{X\sim\{i\}}(E_{x_i}[f|X\sim\{i\}]) = \frac{1}{N} \sum_{j=1}^{N} f(H_j)f(H_j^{(i)})-f_0^2 \quad (A.8) \]

For more details, refer to [29].

**REFERENCES**


