Parameter identification and identifiability analysis of lithium-ion batteries

Yun Young Choi1 ⊗ | Seongyoon Kim1 ⊗ | Kyunghyun Kim1,2 ⊗ | Sanghyun Kim1 ⊗ | Jung-Il Choi1 ⊗

1School of Mathematics and Computing (Computational Science and Engineering), Yonsei University, Seoul, 03722, Republic of Korea
2VOLTWIN Inc., Seoul, Republic of Korea

Correspondence
Jung-II Choi, School of Mathematics and Computing (Computational Science and Engineering), Yonsei University, Seoul, Korea.
Email: jic@yonsei.ac.kr

Funding information
Korea Institute of Science and Technology Information, Grant/Award Number: K-21-SG29-01R-1; National Research Foundation of Korea, Grant/ Award Number: NRF-20151009350; Korea Institute of Energy Technology Evaluation and Planning, Grant/ Award Number: 20214910100070; Seoul Business Agency, Grant/Award Number: SC210026

1 INTRODUCTION

Recently, the demand for rechargeable batteries has increased due to the commercialization of electric vehicles (EVs) as eco-friendly modes of transportation. Lithium-ion batteries (LIBs) are the most widely used power sources for EVs and portable electronics because of their advantages such as light weight, high power, and energy density.1 Nevertheless, battery degradation, fast charging, cell and module optimization, thermal management, and safety are ongoing challenges in researching LIBs.2-6,55

Experimental approaches to address these challenges require high experimental costs originating from the multiphysics and multiscale characteristics of LIBs. Therefore, physics-based models for LIBs, such as equivalent circuit model(ECM) and electrochemical models, have been widely used to tackle such problems.7 The ECMs are ideal for real-time implementation due to their simple structure with lumped parameters.7-12 The electrochemical models,
represented by the pseudo-two-dimensional (P2D) model, can describe electrochemical characteristics inside battery cells with parameters having electrochemical meaning.\textsuperscript{13,14} The P2D model consists of partial differential equations (PDEs) and algebraic equations with multiple model parameters: Butler-Volmer equations, transport equations, and charge conservation equations. The governing equations are defined on the spatial grid in each electrode, membrane, and solid particles at each grid point of an electrode. To develop an electrochemical model, accurate measurements of the model parameters are important because of the reliability and robustness of the LIB model, especially as the number of model parameters increases.\textsuperscript{15} In general, the model parameters are directly measured through additional experiments or calibrations for specific LIBs. However, these approaches require high experimental costs because the various types of LIBs may have different model parameters according to various components, designs, degradation, and even manufacturing processes.\textsuperscript{16}

Alternatively, one may consider utilizing charge/discharge data to identify the electrochemical model parameters based on a genetic algorithm (GA).\textsuperscript{17-21} Forman et al.\textsuperscript{19} introduced a parameter identification (PI) approach to identify 88 parameters of the P2D models from dynamic discharge data using the GA. Zhang et al.\textsuperscript{18} performed a PI using a non-dominated sorting GA to optimize multi-objective functions by using terminal voltage and surface temperature data. PI methods have also been studied using other optimization techniques instead of the GA\textsuperscript{22-26} and a simplified electrochemical model.\textsuperscript{27-29} However, some parameters may not independently affect the model outputs, and they cannot be uniquely determined.\textsuperscript{30} In addition, the negligible effects of the parameters on the model output may contribute to their inaccurate identification. In these cases, even if the model output was well fitted to the experimental data, the identified parameters converged to inadequate values.\textsuperscript{31} Identifiability analysis (IA) addresses the crucial question of whether the reference data are sufficient to identify the model parameters. Various criteria based on the Fisher information matrix (FIM) have been introduced to analyze the identifiability of parameters. For instance, Deng et al.\textsuperscript{20} introduced the ratio between the normalized D and E (RDE) criteria of the FIM to sort out the reduced-order electrochemical model parameters to be identified. In addition, attempts have been made to maximize the parameter identifiability of the LIB model by optimizing the current profile of the reference data for the PI.\textsuperscript{31-35}

Although many studies identify parameters for battery models, there are still issues related to the identifiability and uncertainty analysis of PI methods. First, few studies consider IA from reference data under various operating conditions such as temperature, C-rate, and the range of state of charge (SOC). Since the sample size of data affects the identifiability of model parameters, a criterion independent of the sample size is required for IA under various operating conditions. Second, most PI studies were conducted under either constant current (CC) or dynamic discharge conditions.\textsuperscript{19,20,28,36,37} The data set obtained under dynamic discharge conditions contains the voltage response of the battery to the dynamic current. In contrast, the data set under a single CC discharge condition has limited information about the voltage response of the battery to the single magnitude of the current. However, it is worth noting that the data set obtained under multiple CC discharge conditions contains the voltage response to various static at the same SOC level. Lastly, most PI studies\textsuperscript{19,20,33,38} evaluated the confidential intervals of identified parameters without assessing the uncertainty propagation of the parameters to the voltage responses. However, the uncertainty quantification analysis of model responses might be essential and helpful for reliable prognostics of LIBs based on the modeling approaches.\textsuperscript{39}

In this study, we proposed a PI and IA (PIIA) framework with the sample-averaged RDE criterion to develop

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{PIIA.pdf}
\caption{Schematics of the proposed PIIA of LIB}
\end{figure}
a robust identified-parameter model of the LIB from discharge data under various operating conditions, as shown in Figure 1. We analyzed the CC discharge data under various operating conditions, multiple CC discharge data set and dynamic discharge data. The identifiable parameters from different data sets are compared with respect to the sampled-averaged RDE criterion independent of sample size. The best subset of the model parameters with the best reference data set was determined by maximizing the sample-averaged RDE criterion. Subsequently, we optimized the parameters in the best subset from the data sets based on the GA. The fitness function for the GA was set to the mean squared error of the voltage curves obtained from the electrochemical model. The confidence intervals of the identified parameters were calculated using the Cramér-Rao inequality. Based on the Latin hypercube sampling (LHS), the generated parameters were sampled by confidence intervals of identified parameters. The electrochemical model with generated parameters calculates the empirical distributions of the voltage curves. The output uncertainty of the identified-parameter model was obtained from the bounds of the empirical distribution. This is the first attempt to quantify the uncertainty of the identified-parameter model outputs. The rest of this paper is organized as follows. Section 2 introduces an electrochemical model, the FIM with RDE criterion, Cramér-Rao inequality, GA optimization process, LHS, and details of their implementation. Section 3 presents the results of the identification and validation process by comparing the reference data under various operating conditions. Further, the PI method was also applied to the experimental data under the dynamic stress test conditions (DST) to predict the voltage profile under the federal urban driving schedule (FUDS) cycle in order to validate the robustness of the proposed method. Finally, section 4 presents the conclusions.

2 | METHODOLOGY

2.1 | Governing equation of electrochemical model

The computational domains of the electrochemical model consist of each electrode, separator, and solid particle superimposed on each electrode of the LIB cell, as shown in Figure 2. The electrode and separator are assumed to be one-dimensional in the direction in which the current flows. Solid particles in the electrode are spherical and assumed to be one-dimensional in the radial direction. The Li+ changes in solid particles are predicted by Fick’s second law:

\[
\frac{\partial c_{1,k}}{\partial t} = D_{1,k} \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial c_{1,k}}{\partial r} \right),
\]

where \( c_{1,k} \) is the concentration in the solid phase, and \( D_{1,k} \) is the diffusion coefficient of the solid phase. Note that the subscript \( k = p, s, \) and \( n \), which indicate the positive electrode, separator, and negative electrode, respectively. The variables related to the solid phase are only in each electrode \( k = p, n \). Solving this diffusion equation at every node in the electrode domains increases the computational cost. To reduce the computation time, the concentration distribution in the solid particle is assumed to be quadratic. Then, the average and surface concentrations of the solid particles are obtained as follows:

\[
\frac{\partial c_{1,k}^{avg}}{\partial t} = -3 \frac{j_k}{R_{p,k}},
\]

\[
c_{1,k}^{\ast} - c_{1,k}^{avg} = - \frac{R_{p,k} j_k}{D_{1,k} \sigma^3},
\]

where \( j_k \) is the transfer current density, \( c_{1,k}^{\ast} \) is the concentration at the surface of the solid particle, and \( c_{1,k}^{avg} \) is the average concentration in the solid particle. The lithium-ion concentration in the liquid phase was described by the transport equation, including the diffusion term:

\[
\epsilon_k \frac{\partial c_{2,k}}{\partial t} = \nabla \cdot \left( D_{2,k}^{eff} \nabla c_{2,k} \right) + a_k \left( 1 - \tau_+ \right) j_k,
\]

where \( \epsilon_k \) is the porosity of each electrode, \( \tau_+ \) is the transfer number, \( a_k \) is the specific surface area, and \( D_{2,k}^{eff} \) is the effective diffusion coefficient of the electrolyte. The transfer current densities of each electrode are defined as

\[
j_k = 2k_{eff} \sqrt{c_{2,k} \left( c_{1,k}^{max} - c_{1,k}^* \right)} \frac{c_{1,k}^* \sinh \left( \frac{0.5F}{RT} \eta_k \right)}{k_{eff}},
\]

where \( c_{2,k} \) is the concentration in the liquid phase, \( c_{1,k}^{max} \) is the maximum concentration in the solid particle, and \( I \) is the temperature. \( k_{eff} \) is the effective rate constant, \( R \) is the universal gas constant, and \( F \) is the Faraday’s constant. The overpotential of the electrodes is defined as
\[ \eta_k = \phi_{1,k} - \phi_{2,k} - U_k, \]  
where \( \phi_{1,k} \) and \( \phi_{2,k} \) are the potentials in the solid and liquid phases, respectively. \( U_k \) is the open-circuit potential, which depends on the state of charge at the surface of a solid particle. The charge conservation law to describe the potential in the solid and electrolyte can be expressed as

\[ \nabla \cdot (\kappa^e \nabla \phi_{1,k}) = a_k F j_k, \]  
(7)

\[ \nabla \cdot (\kappa^e \nabla \phi_{2,k}) - \nabla \cdot \left( \kappa^e \frac{2RT}{F} (1 - t^+) \nabla (\ln c_{2,k}) \right) = -a_k F j_k, \]  
(8)

where \( \kappa^e \) is the effective conductivity of the solid phase and \( \kappa^e_k \) is effective electrolyte conductivity.

### 2.2 | Boundary conditions

The boundary condition of the solid phase concentration is defined as

\[ \frac{\partial c_{1,k}}{\partial r} \bigg|_{r=0} = 0, \quad \text{and} \quad \frac{\partial c_{1,k}}{\partial r} \bigg|_{r=R_{p,k}} = -\frac{j_k}{D_{1,k}}. \]  
(9)

At the end of each electrode, the concentration of electrolyte follows the zero-flux condition:

\[ \frac{\partial c_{2,k}}{\partial y} \bigg|_{y=y_p,y_n} = 0. \]  
(10)

Because we assumed a quadratic assumption for the concentration distribution in the solid particles, the boundary condition could not be solved. At the electrode and separator interfaces, the boundary condition follows the continuity of the fluxes. The continuity of the electrolyte concentration condition is defined as:

\[ D^{eff}_{2,k} \frac{\partial c_{2,k}}{\partial y} \bigg|_{y=y_{k_a}} = D^{eff}_{2,k} \frac{\partial c_{2,k}}{\partial y} \bigg|_{y=y_{k_b}}, \quad \text{and} \quad c_{2,k} \bigg|_{y=y_{k_a}} = c_{2,k} \bigg|_{y=y_{k_b}}. \]  
(11)

Note that \( k_a \) and \( k_b \) represent the adjacent domain between each electrode and the separator. For the solid potential, the applied current density \( (I_{app}) \) determined by the operating condition is imposed at the end of each electrode, and the zero-flux condition at the other boundary is as follows:

\[ \sigma^e_k \frac{\partial \phi_{1,k}}{\partial y} \bigg|_{y=y_{p,y_n}} = -I_{app}, \quad \text{and} \quad \sigma^e_k \frac{\partial \phi_{1,k}}{\partial y} \bigg|_{y=y_{p,y_m}} = 0. \]  
(12)

The potential of the electrolyte at the end of the positive electrode and at the end of the negative electrode is as follows:

\[ \kappa^e \frac{\partial \phi_{2,p}}{\partial y} \bigg|_{y=y_p} = 0, \quad \text{and} \quad \phi_{2,n} \bigg|_{y=y_n} = 0. \]  
(13)

The boundary conditions at the interface of the electrode/separator are similar to those of the electrolyte concentration:

\[ \kappa^e \frac{\partial \phi_{2,k}}{\partial y} \bigg|_{y=y_{k_a}} = \kappa^e \frac{\partial \phi_{2,k}}{\partial y} \bigg|_{y=y_{k_b}}, \quad \text{and} \quad \phi_{2,k} \bigg|_{y=y_{k_a}} = \phi_{2,k} \bigg|_{y=y_{k_b}}. \]  
(14)

### 2.3 | Fisher information matrix

A matrix \( \text{FIM} \) includes information about the effect of the variation of model parameters on the variation of the model output, which is defined as

\[ \text{FIM} = \sum_{n=1}^{N} Y_x(n)Q^{-1}_n Y^T_x(n), \]  
(15)

where \( x \) is the vector of \( N_p \) parameters, \( N \) is the total number of experimental or reference data, and \( Q \) is the covariance matrix of the measurement noise. Note that the dimension of \( \text{FIM} \) is \( N \times N \), and the dimension of the covariance matrix is \( N \times N \). \( Y_x \) is the output sensitivity function matrix, which is defined as

\[ Y^T_x(n) = \frac{\partial V_{est,n}(x)}{\partial x^T} \bigg|_{x_0}, \]  
(16)

where \( x \) is the parameter vector, \( V_{est,n}(x) \) is the estimated voltage from the electrochemical model at the sample point, and \( x_0 \) is the reference parameter from the previous research. \(^{41}\)

There are some criteria based on \( \text{FIM} \) to quantify the identifiability of the model parameters for finding the best subset to be identified, such as the D, A, E, and modE criteria. The D criterion is the determinant of the \( \text{FIM} \), which is a value related to the confidence interval of the identified parameter. \(^{42}\) The E criterion is the maximum eigenvalue of \( \text{FIM} \), which is related to the uncertainty in the worst-case direction in the solution space. \(^{43}\) The criterion
of $\text{modE}$ is the ratio between the maximum and minimum eigenvalues of $\text{FIM}$, which is related to whether the influence of the parameter on the model result is independent or not. Recently, the RDE criterion, including the D criterion and $\text{modE}$ criterion, was proposed by Machado et al.\textsuperscript{44} Therefore, we use the RDE criterion for the IA, which is defined as

$$\text{RDE} = \frac{\text{normD}}{\text{modE}} = \frac{D\|x\|^2}{\text{modE}},$$

where $\text{normD}$ is the normalized determinant of $\text{FIM}$, $\text{modE}$ is the ratio between the highest and lowest eigenvalues of $\text{FIM}$, and $\|x\|$ is the Euclidean norm of the parameter vector. The subset with the maximum value of RDE is considered as the best subset, where the parameters in the subset separately affect the model output and have narrower confidence intervals. Therefore, we compared all the possible subsets of all the model parameters to find the best subset with the maximum RDE values.\textsuperscript{38} The confidence intervals of the identified parameters can be calculated based on $\text{FIM}$ with Cramér-Rao bound as follows:\textsuperscript{45}

$$\left[ x_j - t_{a,N-N_p}\sqrt{\left(\text{FIM}^{-1}\right)_{jj}}, x_j + t_{a,N-N_p}\sqrt{\left(\text{FIM}^{-1}\right)_{jj}} \right],$$

where $x_j$ is the $j$th parameter in the best subset, $(\text{FIM}^{-1})_{jj}$ denotes the $j$th diagonal element of the inverse of $\text{FIM}$, and $t$ is the statistical student $t$ with $\alpha = 95\%$ confidence interval. Note that $N - N_p$ denotes the degrees of freedom.

The sample-averaged $\text{FIM}$ was introduced to compare the experimental data under different operating conditions without the effect of the number of available data, as follows:

$$\text{FIM} = \frac{1}{N} \sum_{n=1}^{N} Y(n)Q_n^{-1}Y^T(n).$$

The sample-averaged RDE criterion obtained from the sample-averaged $\text{FIM}$ was defined as the RDE. We assumed that the reference data with the largest sample-averaged RDE were adaptable to perform the PI process among the reference data under various operating conditions.

### 2.4 Identification process based on genetic algorithm

The GA allows us to find a solution robustly without converging on the local solution.\textsuperscript{46} The GA finds a vector of optimal parameters $x_{\text{opt}}$ such that

$$F(x_{\text{opt}}) \leq F(x); \forall x \in \Omega,$$

where $F$ is the fitness function and $\Omega$ is the solution space, which contains every possible solution of $x$. The candidate solution vector is considered to be a chromosome and undergoes genetic operations inspired by natural genetic systems.\textsuperscript{47} The genetic operation consists of selection, crossover, elitism, and mutation operations to find the optimum vector. The candidate solution at $k$th generation is defined as $x_m^{(k)} (m = 1, 2, \ldots, M)$. Here, $M$ is the population size.

In previous PI studies, there were different model parameters to be identified.\textsuperscript{17-23.27-29} In this study, we exclude parameters related to the material of each electrode and the geometry of the LIB cell from the model parameters to be identified: the transference number, open-circuit voltage, maximum concentration in the solid phase, diffusion coefficient of the solid phase, and conductivity of each electrode. We included all the remaining 33 parameters in the electrochemical model to find the best subset to be identified: the specific surface ($a_p, a_n$), solid particle radius ($R_{p, p}, R_{p, n}$), porosity ($\epsilon_p, \epsilon_n$), filler fraction ($\epsilon_{2, p}, \epsilon_{2, n}$), rate constant ($k_p, k_n$), diffusion coefficient of the electrolyte ($D_{2, p}, D_{2, n}$), Bruggeman’s coefficient ($\text{brugg}$), constant for the conductivity equation of the electrolyte ($\theta$), initial solid phase concentration ($c_0^{0,1, p}, c_0^{0,1, n}$), initial electrolyte concentration ($c_0^0$), and activation energy of each parameter ($E_a^0, E_a^{1, p}, E_a^{1, n}, E_a^{2, p}, E_a^{2, n}, E_a^{3, p}, E_a^{3, n}$). Note that the subscripts $p, s,$ and $n$ indicate the positive electrode, separator, and negative electrode, respectively. Here, the conductivity of the electrolyte is expressed as follows:

$$\kappa_{\text{eff}} = \kappa_k^{\text{brugg}} \left( \theta(1) + \theta(2)c_{2, k} - \theta(3)c_{2, k}^2 + \theta(4)c_{2, k}^3 - \theta(5)c_{2, k}^4 \right),$$

where $\theta(j)$ is the coefficient of electrolyte conductivity. The Arrhenius equation is used to describe the temperature dependence of the model parameter as follows:\textsuperscript{28}

$$\psi = \psi_{\text{ref}} \exp \left[ \frac{E_a^\psi}{R} \left( \frac{1}{T_{\text{ref}}} - \frac{1}{T} \right) \right],$$

where $\psi_{\text{ref}}$ indicates the temperature-dependent parameters ($\kappa_k, D_{1, k}, \kappa_{\text{eff}}, D_{2, k}, E_a^\psi$). $E_a^\psi$ is the activation energy of the parameter $\psi_{\text{ref}}$.

The fitness function in the GA process is based on the root mean squared error between the reference data and the model output, and it is expressed as follows:

$$F(x_m^{(k)}, V_{\text{obs}}) = \frac{1}{N} \sum_{n=1}^{N} \left( V_{\text{obs}, n} - V_{\text{est}, n}(x_m^{(k)}) \right)^2,$$
2.5 | Latin hypercube sampling

The LHS method is an efficient probabilistic sampling method for the propagation of uncertainty due to its better convergence compared to that of the standard Monte Carlo (MC) method. Therefore, we efficiently calculate the uncertainty of the model outputs based on the confidence intervals of the identified parameters to validate the robustness of the identified-parameter model. First, \( N_s \) sets of identifiable parameters were uniformly generated using the LHS. The generated identifiable parameters with reference values of the unidentifiable parameters can be written as:

\[
\mathbf{x}_{n_s} = \left( x_{n_s,1}, x_{n_s,2}, \ldots, x_{n_s,J}, \hat{x}_1, \hat{x}_2, \ldots, \hat{x}_K \right), \quad n_s = 1, 2, \ldots, N_s,
\]

where \( x_{n_s,j} \) is a generated value of the \( j \)th identifiable parameter for \( j = 1, 2, \ldots, J \) and \( \hat{x}_k \) is a reference value of the \( k \)th unidentifiable parameter for \( k = 1, 2, \ldots, K \). Using the generated parameter set \( \mathbf{x}_{n_s} \) for \( n_s = 1, 2, \ldots, N_s \), the empirical distribution of the voltage curve is estimated as \( \{ V_{est}(\mathbf{x}_{n_s}) \}_{n_s=1}^{N} \). From the estimated empirical distribution, the upper and lower bounds of the voltage can be obtained.

2.6 | Details of implementation

We used a finite difference method on a staggered grid to solve the electrochemical model in our PI method. The transport equations were discretized by the Crank-Nicolson scheme in time, and the spatial derivative terms in the governing equations were discretized by the second-order central-difference scheme. The output sensitivity matrix \( Y^T_x(n) \) for obtaining the Fisher information matrix (FIM) is expressed as:

\[
Y^T_x(n) \approx \frac{V_{est,n}(\mathbf{x} + \Delta \mathbf{x}) - V_{est,n}(\mathbf{x} - \Delta \mathbf{x})}{2\Delta \mathbf{x}}, \quad \text{for} \quad \mathbf{x} \in \Omega.
\]

where \( \Delta \mathbf{x} \) is the perturbation factor, which is assumed to be 0.1% of the value of each parameter. Note that we assume that the measurement error does not exceed 0.01% of the sample data. The function \( \text{ga} \) in the global optimization toolbox of MATLAB® 2018 was used for the optimization process to find the optimal parameters in the best subset. The parameters and functions of the GA process are summarized in Table 1. GA optimization iteratively performs genetic operations, until the relative change in the minimum fitness converges to the convergence criterion during consecutive \( R_0 \) generations as follows:

\[
\frac{1}{R_0} \min_{k' \in [k-R_0,k]} \left( \min_{m} F(x^{(k')}_m) \right) \leq \epsilon_{tol}
\]

where \( R_0 \) is the maximum stall generation number and \( \epsilon_{tol} \) is the tolerance.

3 | RESULTS AND DISCUSSION

3.1 | Validation of electrochemical model

We validated the electrochemical model by comparing the results with those of a well-known open-source P2D model solver, the Li-ion simulation battery toolbox (LIONSIMBA). We simulated the case of a constant current (CC) discharge under isothermal conditions with a temperature of 298.15 K, 1 C of 29.5 A m\(^{-2}\), and a cutoff voltage of 2.5 V. The model parameters and operating conditions used for the validation are listed in Table 2. The

| TABLE 1 Summary of parameters and functions used in the present GA |
|---------------------------------------------------------------|
| Description | Function or value |
| Initial parameter set | Uniform |
| Population size, \( M \) | 80 |
| Selection operation | Stochastic Uniform Function |
| Crossover operation | Intermediate Function with a fraction of 0.8 |
| Scaling method | Rank |
| Elite number | 2 |
| Maximum stall generation number, \( R_0 \) | 50 |
| Tolerance, \( \epsilon_{tol} \) | \( 10^{-6} \) |

where \( N \) is the number of data points in the voltage curves, and \( n \) indicates a sample point with different time or SOC values. \( V_{est,n}(\mathbf{x}^{(k)}_m) \) is the estimated voltage from the electrochemical model with a given parameter vector \( \mathbf{x}^{(k)}_m \), and \( V_{obs,n} \) is the observed voltage from the experimental or sample data.
coefficients calculated by using the variables and other constants are listed in Table 3.

Before proceeding to validation, we performed spatial and temporal grid convergence tests with five different grid resolutions (16, 32, 64, 128, and 256 points for each domain) and four different time intervals (100, 10, 5, and 1 s for each time step). Considering the computational costs and accuracy of the solution, we used 32 grids for each domain (positive electrode, separator, and negative electrode) as the baseline grid resolution with 5 s intervals. Figure 3 indicates that the cell voltages obtained from the present model are consistent with the results in reference49 under 0.5C, 1C, and 2C discharge; the mean relative differences are less than 0.2%.

Figure 4A shows the variation in the average solid phase concentration in each domain by changing the C-rate from 0.5C to 2C at time $t = 1000$ s. In the negative electrode, the solid phase concentration near the separator approached zero. As the discharge current increased, the solid phase concentration showed a more significant variation. Figure 4B shows the change in electrolyte concentration in each domain by changing the C-rate from 0.5C to 2C at time $t = 1000$ s. The electrolyte concentration decreased in the positive electrode as the C-rate increased, whereas the electrolyte concentration increased in the negative electrode. The change in electrolyte concentration is less than 30% in each electrode at the 0.5C-rate discharge. However, the change in electrolyte concentration is more than 70% at the 2C-rate discharge near the end of the positive electrode. We confirmed that the concentration distribution was similar to that reported in a previous study.52

### 3.2 Parameter identification with identifiability analysis

The proposed PIIA framework was applied to reference data calculated from the electrochemical model with the reference parameters in the previous section under

| Symbol   | Description [Unit] | PE          | Separator | NE          |
|----------|--------------------|-------------|-----------|-------------|
| $l_k$    | Thickness of electrode [m] | $8 \times 10^{-5}$ | $2.5 \times 10^{-5}$ | $8.8 \times 10^{-5}$ |
| $R_p$    | Radius of Solid particle [m] | $2 \times 10^{-6}$ | —         | $2.0 \times 10^{-6}$ |
| $\sigma_k$ | Conductivity of electrode [sm$^{-1}$] | 100         | —         | 100         |
| $t_+$    | Transference number | 0.364       | 0.364     | 0.364       |
| $D_{1,k}$ | Diffusion coefficient of solid phase | $1.0 \times 10^{-14}$ | —         | $3.9 \times 10^{-14}$ |
| $c_{\text{max}}^k$ | Maximum solid phase concentration | 51554       | —         | 30555       |
| $d_p$    | Specific surface area [m$^2$m$^{-3}$] | 885000      | —         | 723600      |
| $e_k$    | Porosity           | 0.385       | 0.724     | 0.485       |
| $e_{2,k}$ | Filler fraction | 0.025       | —         | 0.0326      |
| $k_{2,k}$ | Rate constant [m$^2$ mol$^{-0.5}$ s$^{-1}$] | $2.334 \times 10^{-11}$ | —         | $5.031 \times 10^{-11}$ |
| $D_{2,k}$ | Diffusion coefficient of electrolyte [m$^2$s$^{-1}$] | $7.5 \times 10^{-10}$ | $7.5 \times 10^{-10}$ | $7.5 \times 10^{-10}$ |
| $c_{0,1,k}$ | Initial solid phase concentration [mol m$^{-3}$] | 25751       | —         | 26128       |
| $E_a^k$  | Reaction constant activation energy [J mol$^{-1}$] | 5000        | —         | 5000        |
| $E_a^{D_1,k}$ | Diffusion coefficient of solid phase activation energy [J mol$^{-1}$] | 5000      | —         | 5000        |
| $E_a^{D_2,k}$ | Electrolyte conductivity activation energy [J mol$^{-1}$] | 5000  | 5000     | 5000        |
| $E_a^{D_3,k}$ | Diffusion coefficient of electrolyte activation energy [J mol$^{-1}$] | 5000  | 5000     | 5000        |
| brugg   | Bruggeman’s coefficient | 4           |           |             |
| $\theta(1)$ | Constant for conductivity equation of electrolyte | 0.0413     |           |             |
| $\theta(2)$ | Constant for conductivity equation of electrolyte | $5 \times 10^{-4}$ |           |             |
| $\theta(3)$ | Constant for conductivity equation of electrolyte | $4.72 \times 10^{-7}$ |           |             |
| $\theta(4)$ | Constant for conductivity equation of electrolyte | $1.509 \times 10^{-10}$ |           |             |
| $\theta(5)$ | Constant for conductivity equation of electrolyte | $1.602 \times 10^{-14}$ |           |             |
| $c_{0}^e$ | Initial concentration in the electrolyte [mol m$^{-3}$] | 1000        |           |             |
TABLE 3 Open-circuit potential and other coefficients, which are calculated by variables of the electrochemical model⁴⁹

| Description                                   | Relationship or Empirical formula                                                                 |
|-----------------------------------------------|-----------------------------------------------------------------------------------------------------|
| Open-circuit potential                        | \(U_{1,rel} = \frac{-4.658 + 83.6692\delta_n^0 - 401.11968\delta_n^0 + 342.9098\delta_n^0 - 462.4718\delta_n^0 + 433.1436\delta_n^0}{-1 + 18.9339\theta_n + 79.3352\theta_n + 37.3111\theta_n + 73.0808\theta_n + 95.9669\delta_n^0} \) |
|                                              | \(U_{2,rel} = 0.7222 + 0.1387\theta_n + 0.029\theta_n^3 - 0.0172/\theta_n + 0.0019/\theta_n^3 + 0.2808\exp(0.9 - 15\theta_n) - 0.7984\exp(0.4465\theta_n - 0.4108) \) |
|                                              | \(\theta_k = c_{1,k}^{c,\text{max}} \)                                                                 |
| Entropy change                                | \(\frac{\Delta S}{\Delta T}\text{ (rel)} = -0.001 \left( \frac{a_{\theta_n}^0 + a_{\theta_n}^1\theta_n + a_{\theta_n}^2\theta_n + a_{\theta_n}^3\theta_n}{1 + a_{\theta_n}^0 + a_{\theta_n}^1\theta_n + a_{\theta_n}^2\theta_n + a_{\theta_n}^3\theta_n} \right) \) |
|                                              | \(a_{\theta_n}^0 = 0.199521039, a_{\theta_n}^1 = -0.928373822, a_{\theta_n}^2 = 1.36455068900003, a_{\theta_n}^3 = -0.611544893999998 \) |
|                                              | \(a_{\theta_n}^4 = -5.66147988699997, a_{\theta_n}^5 = 11.47636191, a_{\theta_n}^6 = -9.824312135999998, a_{\theta_n}^7 = 3.048755063 \) |
|                                              | \(\frac{\Delta S}{\Delta T}\text{ (rel)} = 0.001 \left( \frac{a_{\theta_n}^0 + a_{\theta_n}^1\theta_n + a_{\theta_n}^2\theta_n + a_{\theta_n}^3\theta_n + a_{\theta_n}^4\theta_n + a_{\theta_n}^5\theta_n + a_{\theta_n}^6\theta_n + a_{\theta_n}^7\theta_n}{1 + a_{\theta_n}^0 + a_{\theta_n}^1\theta_n + a_{\theta_n}^2\theta_n + a_{\theta_n}^3\theta_n + a_{\theta_n}^4\theta_n + a_{\theta_n}^5\theta_n + a_{\theta_n}^6\theta_n + a_{\theta_n}^7\theta_n} \right) \) |
|                                              | \(a_{\theta_n}^0 = 0.005269056, a_{\theta_n}^1 = 3.299265709, a_{\theta_n}^2 = -91.79325798, a_{\theta_n}^3 = 1004.911008, a_{\theta_n}^4 = -5812.278127 \) |
|                                              | \(a_{\theta_n}^5 = 19329.7549, a_{\theta_n}^6 = -37147.8947, a_{\theta_n}^7 = 38379.18127, a_{\theta_n}^8 = -16515.05308 \) |
|                                              | \(a_{\theta_n}^9 = -48.09287227, a_{\theta_n}^{10} = 1017.234804, a_{\theta_n}^{11} = -10481.80419, a_{\theta_n}^{12} = 59431.3 \) |
|                                              | \(a_{\theta_n}^{13} = -195881.6488, a_{\theta_n}^{14} = 374577.3152, a_{\theta_n}^{15} = -385821.1607, a_{\theta_n}^{16} = 165705.8597 \) |
| Diffusion coefficient                         | \(D_2^{\text{eff}} = D_{2,k}^{\text{brug}} \)                                                                 |
| Effective electrolyte conductivity            | \(k_k^{\text{eff}} = k_k^{\text{brug}}\left(4.1253 \times 10^{-2} + 5.007 \times 10^{-4}c_2k - 4.712 \times 10^{-7}c_2k^2 + 1.5094 \times 10^{-10}c_2k^3 - 1.6018 \times 10^{-14}c_2k^4\right)\) |
| Effective electrode conductivity              | \(\sigma_k^{\text{eff}} = \sigma_k(1 - \epsilon_k - \epsilon_f,k) \)                                                                 |

FIGURE 3 Comparisons of cell voltages in reference⁴⁹ and obtained from the present electrochemical model under 0.5C, 1C, and 2C discharge

various operating conditions (OC): three temperature conditions (273.15, 298.15, and 318.15 K), three C-rate conditions (0.5C, 1C, 2C), three SOC ranges (narrow, medium, and full), and two dynamic discharge conditions (DST and FUDS). Here, each SOC range is [1500–2500 s], [500–3100 s] and [0–3600 s], respectively. Note that the baseline operating condition (OC1) is 1C and 298.15 K with the full range in the SOC, and multiple CC data sets (OC8, OC9, and OC10) contain three or more CC data sets with different operating conditions.

Table 4 shows the number of parameters included in the best subset, the maximum value of sample-averaged RDE, and the parameters in the best subsets obtained from each reference data set under various operating conditions. We confirmed that the number of parameters in the best subset and the values of the sample-averaged...
RDE depended on the data sets. We first verified that the
given data set’s discharge rate and SOC range change the
number of identified parameters and the magnitude of
sample-averaged RDE. By contrast, the temperature did
not significantly affect the identifiability of the
parameters. The number of parameters in most of the single CC
data sets is similar to that obtained from previous PI re-
searches. Although the number of parameters in the
best subset is similar for single and multiple CC data sets,
we observed that using the multiple CC data sets increases
the parameter identifiability compared to using a single
data set. Besides, the value of sample-averaged RDE sig-
ficantly increases even when compared with the identi-
fiability of the data set under the DST and FUDS
conditions. The RDE, as a representative of the uncer-
tainty of identified parameters, demonstrates the im-
proved quality of these parameters from using multiple
CC data sets under various operating conditions. These
results indicate that collecting data under various oper-
ating conditions is necessary for robust PI to increase the
identifiability of the identified parameters. The rate con-
stant (k) is included in the best subset of the reference data
under every OC. In most cases of OC, the particle radius
($R_{p,k}$), constants of electrolyte conductivity ($\theta(k)$), and
diffusion coefficient of the electrolyte ($D_{2,k}$) are included in
the best subset. However, in terms of the RDE criterion,
the specific surface area ($a_{S}$) and parameter related to the
activation energy ($E_{a,k}$, $E_{a,k}^R$, $E_{a,k}^p$, and $E_{a,k}^{2,s}$) are not
included in every best subset.

We chose the reference data under OC10 for the best
identified-parameter model and the reference data under
OC7 for the worst identified-parameter model in terms of
the value of sample-averaged RDE. Figure 5 shows the
maximum sample-averaged RDE at data sets depending
on the number of parameters in a subset. Note that the

| Operating condition | $N_{max}$ | $\log_{10}RDE_{max}$ | Identifiable parameters |
|---------------------|-----------|----------------------|------------------------|
| OC1 1C, 298.15K     | 14        | 191.0                | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $D_{2,n}$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |
| OC2 0.5C, 298.15K   | 14        | 188.0                | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $D_{2,n}$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |
| OC3 2C, 298.15K     | 10        | 115.6                | $e_{p}$, $e_{n}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(1)$, $\theta(2)$, $\theta(4)$, $\theta(5)$ |
| OC4 1C, 318.15K     | 14        | 191.1                | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |
| OC5 1C, 278.15K     | 14        | 191.5                | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |
| OC6 1C, 298.15K, [500s, 3100s] | 9 | 151.3 | $R_{p,n}$, $k_{n}$, $D_{2,n}$, $D_{2,n}$, $\theta(4)$, $\theta(5)$ |
| OC7 1C, 298.15K, [1500s, 2500s] | 9 | 113.2 | $R_{p,p}$, $R_{p,n}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(4)$, $\theta(5)$ |
| OC8 OC1, OC2, and OC3 | 15 | 215.7 | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(1)$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |
| OC9 OC1, OC4, and OC5 | 15 | 201.0 | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(1)$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |
| OC10 OC1, OC2, OC3, OC4, and OC5 | 15 | 216.0 | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $e_{2,p}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(1)$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |
| DST DST, 298.15K     | 16        | 194.4                | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $e_{2,p}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |
| FUDS FUDS, 298.15K   | 16        | 190.7                | $R_{p,p}$, $R_{p,n}$, $e_{p}$, $e_{n}$, $e_{2,p}$, $k_{n}$, $D_{2,p}$, $D_{2,n}$, $\theta(2)$, $\theta(3)$, $\theta(4)$, $\theta(5)$, $\epsilon_{1,n}^{c}$ |

| FIGURE 5 | Comparisons of sample-averaged RDE curves from reference data under each operating condition |                     |

negative values of $\log_{10}RDE$ were set to 0 to clarify the
display. The RDE values at OC1, OC7, DST, and FUDS data
set remained at zero for more than 24 parameters. This is
because by Eq. (21), the influence of the parameters rel-
ted to the activation energy on the model output reduces
to zero. Conversely, the RDE curve at OC10 has some val-
ues for the parameters related to the activation energy due
to the OC10, including the reference data under different
 temperatures. The results of the IA indicate that the best
subset with maximum sample-averaged RDE consists of
15 identifiable parameters, which are listed in Table 5.

We performed the GA process until the minimum
values of the fitness function satisfied the convergence
criterion. The minimum values of the fitness function
converted to 0.0394 at OC10 and 0.000798 at OC7. The opti-
mal parameters with the minimum values of the fitness
function in each case are shown in Table 5. The identified
parameters at OC10 are of the same order of magnitude
or even close to the reference parameters. Although the
identified parameters related to the electrolyte conductivity
have wide confidence intervals, most of the parameters
| Input parameter | Value OC10 | CI [Min, Max] OC10 | Value OC11 | CI [Min, Max] OC11 | Reference |
|----------------|-----------|----------------|-----------|----------------|-----------|
| Specific surface area (Positive) $A_s$ | 885000 | — | 25999 | 26008 | — | |
| Specific surface area (Negative) $A_{s,n}$ | 723600 | — | 667 | 75 | — | |
| Particle radius (Positive) $r_p$ | 2.0 × 10^{-4} | — | 1.0 × 10^{-6} | 2.0 × 10^{-6} | 1.82 × 10^{-6} | 3.148 |
| Particle radius (Negative) $r_{p,n}$ | 2.0 × 10^{-4} | — | 1.0 × 10^{-6} | 2.0 × 10^{-6} | 1.82 × 10^{-6} | 3.148 |
| Porosity (Positive) $\phi_p$ | 0.724 | — | 0.185 | 0.381 | 0.380 | 0.381 |
| Porosity (Negative) $\phi_{p,n}$ | 0.485 | — | 0.285 | 0.685 | 0.432 | 0.434 |
| Filler fraction (Positive) $\phi_f$ | 0.025 | — | 0.025 | 0.025 | 0.025 | 0.025 |
| Filler fraction (Negative) $\phi_{f,n}$ | 0.032 | — | 0.032 | 0.032 | 0.032 | 0.032 |
| Rate constant (Positive) $k_1$ | 2.34 × 10^{-11} | — | 9.336 × 10^{-11} | 3.546 × 10^{-11} | 2.99 × 10^{-10} | 6.13 |
| Rate constant (Negative) $k_{1,n}$ | 2.34 × 10^{-11} | — | 9.336 × 10^{-11} | 3.546 × 10^{-11} | 2.99 × 10^{-10} | 6.13 |
| Diffusion coefficient of electrolyte $D_p$ | 7.5 × 10^{-6} | — | 1.875 × 10^{-6} | 1.875 × 10^{-6} | 1.875 × 10^{-6} | 1.875 |
| Diffusion coefficient of electrolyte $D_{p,n}$ | 7.5 × 10^{-6} | — | 1.875 × 10^{-6} | 1.875 × 10^{-6} | 1.875 × 10^{-6} | 1.875 |
| Diffusion coefficient of electrolyte (Positive) $D_{p,+}$ | 7.5 × 10^{-6} | — | 1.875 × 10^{-6} | 1.875 × 10^{-6} | 1.875 × 10^{-6} | 1.875 |
| Diffusion coefficient of electrolyte (Negative) $D_{p,-}$ | 7.5 × 10^{-6} | — | 1.875 × 10^{-6} | 1.875 × 10^{-6} | 1.875 × 10^{-6} | 1.875 |
| Bruggeman's coefficient $\alpha$ | — | — | 0.025 | 0.025 | 0.025 | 0.025 |
| Initial solid phase concentration $C_0$ | — | — | — | 0.00031 | 0.00031 | 0.00031 |

**Table 5:** Summary of the reference model parameters and identified parameters for the LIB obtained from reference data at OC10 and OC7. Note that $R_{CI}$ is a relative half of confidence intervals to each parameter.
| Symbol | Description                                      | Unit     | Reference | Input range [Min, Max] | Value (OC10) | CI [Min, Max] (OC10) | $R_{CI}$ (%) | Value (OC7) |
|--------|--------------------------------------------------|----------|-----------|------------------------|--------------|----------------------|--------------|-------------|
| $c_2^0$ | Initial concentration in the electrolyte        | mol m$^{-3}$ | 1000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{R}$ | Reaction constant activation energy (Positive) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{c}$ | Reaction constant activation energy (Negative)  | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{D_s}$ | Diffusion coefficient of solid phase activation energy (Positive) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{D_n}$ | Diffusion coefficient of solid phase activation energy (Negative) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{u_1D_705}$ | Electrolyte conductivity activation energy (Positive) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{u_1D_s}$ | Electrolyte conductivity activation energy (Separator) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{u_1D_n}$ | Electrolyte conductivity activation energy (Negative) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $-E_{a}^{D_s}$ | Diffusion coefficient of electrolyte activation energy (Positive) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{D_s}$ | Diffusion coefficient of electrolyte activation energy (Separator) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
| $E_{a}^{D_n}$ | Diffusion coefficient of solid phase activation energy (Negative) | J mol$^{-1}$ | 5000      | —                      | —            | —                    | —            | —           |
were identified with good accuracy ($R_{CI} < 10\%$) from the reference data under OC10. The identified parameters from the reference data under OC7 were also in a similar order of magnitude to the reference parameter values, but the confidence intervals could not be calculated because of inverse of the FIM involving complex numbers. This is because the reference data under OC7 were insufficient to identify 15 parameters, as shown in Figure 5.

Figure 6 illustrates the comparison of discharge voltage curves from the reference data between the worst identified-parameter model from the reference data under OC7 and the best identified-parameter model from the reference data under OC10. In the best identified-parameter model, the identified voltage curves are consistent with the reference data and have a mean relative error less than 0.2%. Although the worst identified-parameter model adequately estimates the voltage curves in the initial region of the discharge operation, the voltage curves deviate from the reference data as the discharging process continues. This is because the worst identified-parameter model is obtained from the reference data under the narrow SOC range. We confirmed that even if the fitness function sufficiently converges and the scales of the identified parameter are similar to the reference parameter, the identified parameter may be inappropriate to represent the discharge characteristics of an LIB.

Further, we compared the model results with the reference data under the operating conditions for validation cases (OC11, OC12, OC13, OC14, DST, and FUDS), which are listed in Table 6. Figure 7 shows the reference data, voltage curves obtained from the worst identified-parameter model, and voltage curves with voltage uncertainty obtained from the best identified-parameter models. The best identified-parameter model sufficiently predicts the voltage curves with mean relative errors of 0.648%, 0.329%, 0.0888%, 0.145%, 0.315%, and 0.347%, respectively. Although the uncertainty in voltage response is relatively significant at a high C-rate condition compared to low C-rate and dynamic discharge conditions, the uncertainty of identified-parameter models obtained from multiple CC data sets is sufficiently small to predict battery voltage responses accurately. The worst identified-parameter model could not adequately predict the voltages in Figure 7A and D, while the converged fitness value and errors in some identification results are sufficiently small. It seems that PI without IA could not identify the parameters to represent the discharge characteristics of an LIB cell under various operating conditions. The errors in the validation results are larger than the errors in the identification results; however, the best identified-parameter model predicts reasonable voltage curves with narrow uncertainty bounds than those obtained from the worst identified-parameter model. Table 6 summarizes the root mean square error (RMSE) of the identification and validation results obtained from the best identified-parameter model. The values of RMSE and error bounds were sufficiently small in both the identification and validation results.

### 3.3 Parameter identification for real-world problem

To validate the robustness of the proposed PIIA framework, we applied the present framework to a real-world problem and constructed an electrochemical model for the INR 18650-20R cylindrical LIB cell experimental data under dynamic stress test (DST). We validated the identified-parameter model using the federal urban driving schedule (FUDS) cycle data. The experimental data were obtained from the Center for Advanced Life Cycle
TABLE 6  RMSE values for identified-parameter model (OC10). Here, the mean square errors are calculated by the voltage data larger than the cutoff voltage value (2.5 V)

| Identification | Operating conditions | RMSE   |
|----------------|----------------------|--------|
| OC1            | 1C, 298.15K          | 0.0062 |
| OC2            | 0.5C, 298.15K        | 0.0158 |
| OC3            | 2C, 298.15K          | 0.0052 |
| OC4            | 1C, 318.15K          | 0.0052 |
| OC5            | 1C, 278.15K          | 0.0082 |

| Validation     | Operating conditions | RMSE   |
|----------------|----------------------|--------|
| OC11           | 4C, 298.15K          | 0.0246 |
| OC12           | 0.25C, 298.15K       | 0.0243 |
| OC13           | 1C, 323.15K          | 0.0051 |
| OC14           | 1C, 273.15K          | 0.0089 |
| DST            | DST, 273.15K         | 0.0218 |
| FUDS           | FUDS, 273.15K        | 0.0237 |

FIGURE 7  Comparisons of cell voltages from reference data and obtained from identified-parameter models (PI models) under (A) OC11, (B) OC12, (C) OC13, (D) OC14, (E) DST, and (F) FUDS. Here, the shaded region indicates uncertainty bounds of model outputs.
The present LIB cell with a nominal discharge capacity of 2000 mAh consists of a lithium-nickel-manganese-cobaltoxide (LNMC) cathode and a graphite anode. The experimental data for both DST and the FUDS cycles were operated at 298.15 K for 80% battery level. We identified the model parameters in the best subset through experimental data under the DST cycle. We then validated the identified-parameter model by comparing the predicted voltage curve based on the identified parameters from the DST and the experimental data under the FUDS cycle, which was assumed to be the EV operation. Unlike the case in the previous section, there are some unknown geometry factors, such as the thickness and area of each electrode and separator. Therefore, these parameters should be included in the optimal subset. The maximum concentrations of lithium-ion, diffusion coefficients of the solid phase, and the OCV of each electrode were determined by referring to the values provided by the COMSOL built-in setting. The reference values of the initial solid phase concentration in the negative electrode and the initial concentration in the electrolyte were also modified based on the information given by the CALCE battery research group. We used the present electrochemical model under isothermal conditions and 32 grids for each domain because of the computational cost. Here, the initial SOC was assumed to be 80%. The total sample size for the PI was 10660 and 11103 for the experimental data under DST and FUDS, respectively. The parameters in the GA process were the same as those described in the previous section. We performed the PI process with 19 parameters until the minimum value of the fitness function converged to 0.0245.

Table 7 lists the values of the identified parameter for the INR 18650-20R battery. The values of the identified parameters are similar in magnitude to those of the reference data in the previous section. We also confirmed that the values of the identified parameters related to the cell geometry were similar to those in the previous studies. Although most of the parameters in the best subset were identified accurately ($R_{CI} < 10\%$), the confidence intervals were relatively wider than those in Section 3.2. Since the best subset of the identified parameters contains electrode geometry parameters closely related to other parameters, the correlated features of the parameters may increase their confidential intervals in the PI process.

Figure 8A shows that the voltage curves obtained from the identified-parameter model are in good agreement with those from the experimental data during the DST, with a mean relative error of 0.38%. Figure 8B shows a comparison of the experimental data and voltage curves with uncertainty obtained from the identified-parameter model under the FUDS. The mean relative error between the experimental data and the results of the identified-parameter model was 0.46%. Although the uncertainty bounds of outputs are relatively wide compared to those in the previous section, most of the experimental data lie within the uncertainty bounds.

4 | CONCLUSION

We proposed a PIIA framework to develop a robust electrochemical model for LIBs. First, we validated the electrochemical model by comparing it with the reference results. We analyzed the identifiability of the electrochemical model parameters based on the reference data under 12 operating conditions. The present PIIA indicated that the SOC range of the operating conditions was most influential in achieving a robust PI, and the sample-averaged RDE of multiple CC data sets was higher than that of a single CC data set. The multiple CC data sets with the maximum value of the sample-averaged RDE were determined to identify 15 model parameters in the best subset. The parameters in the best subset were identified by the GA process with the fitness function as the mean squared error of the voltage between the reference data and the model output of the parameter subset. The values of the identified parameters were similar to those used in the reference data, with a narrow confidence interval ($R_{CI} < 10\%$) calculated by the Cramér–Rao inequality. Using the LHS, the generated parameters were sampled based on the confidential intervals of the identified parameters. Further, we obtained the empirical distribution of the voltage curves by solving the electrochemical model with the generated parameters and estimated uncertainty bounds of identified identified-parameter model. Although the converged fitness value of narrow SOC data (0.000798) is smaller than that of multiple CC data sets (0.0394), the identified-parameter model from multiple CC data sets accurately predicted voltage responses with narrow uncertainty bounds under the operating conditions for various validation cases within 1% mean relative error. Furthermore, we examined the robustness of the proposed PI method by applying experimental data under the DST and FUDS cycles. The identification and validation results are consistent with the experimental data, with mean relative errors of less than 0.46%. We demonstrated that the identified-parameter model obtained from the proposed identifiability analysis and the PI method represents the discharge characteristics of an LIB. A limitation of the present study is that the electrochemical model does not include thermal models. The absence of the thermal model may limit the effect of the parameters on the model outputs, making it difficult to identify the parameters. In our future work, we will apply identifiability analysis and the PI method to experimental data including temperature.
Table 7 Summary of reference model parameters and identified parameters for the LIB cell under the DST cycle. Note that $R_C$ is a relative half of confidence intervals to each parameter and the superscript * indicates a reference value for the unidentified parameters

| Symbol | Description | Unit | Input range [Min, Max] | Value | CI [Min, Max] | $R_C$ (%) |
|--------|-------------|------|-------------------------|-------|--------------|-----------|
| $a_p$  | Specific surface area (Positive) | $m^2 m^{-3}$ | — | — | 885000* | — | — |
| $a_n$  | Specific surface area (Negative) | $m^2 m^{-3}$ | — | — | 723600* | — | — |
| $R_{p,p}$  | Particle radius (Positive) | $m$ | $[2.0 \times 10^{-7}, 2.0 \times 10^{-5}]$ | $1.91 \times 10^{-6}$ | $[1.89 \times 10^{-6}, 1.93 \times 10^{-6}]$ | 1.08% |
| $R_{p,n}$  | Particle radius (Negative) | $m$ | $[2.0 \times 10^{-7}, 2.0 \times 10^{-5}]$ | $2.98 \times 10^{-6}$ | $[2.96 \times 10^{-6}, 3.01 \times 10^{-6}]$ | 0.83% |
| $\epsilon_p$  | Porosity (Positive) | — | $[0.1, 0.9]$ | 0.22 | $[0.225, 0.230]$ | 1.08% |
| $\epsilon_s$  | Porosity (Separator) | — | $[0.1, 0.9]$ | 0.724* | — | — |
| $\epsilon_n$  | Porosity (Negative) | — | $[0.1, 0.9]$ | 0.470 | $[0.463, 0.477]$ | 1.44% |
| $\epsilon_{2,p}$  | Filler fraction (Positive) | — | — | 0.025* | — | — |
| $\epsilon_{2,n}$  | Filler fraction (Negative) | — | — | 0.0326* | — | — |
| $k_p$  | Rate constant (Positive) | $m^{2.5} \text{ mol}^{-0.5} \text{s}^{-1}$ | $[2.334 \times 10^{-12}, 2.334 \times 10^{-10}]$ | $2.36 \times 10^{-11}$ | $[2.34 \times 10^{-11}, 2.39 \times 10^{-11}]$ | 1.08% |
| $k_n$  | Rate constant (Negative) | $m^{2.5} \text{ mol}^{-0.5} \text{s}^{-1}$ | $[5.031 \times 10^{-12}, 5.031 \times 10^{-10}]$ | $3.79 \times 10^{-11}$ | $[3.74 \times 10^{-11}, 3.84 \times 10^{-11}]$ | 1.20% |
| $D_{2,p}$  | Diffusion coefficient of electrolyte (Positive) | $m^2 \text{s}^{-1}$ | $[7.5 \times 10^{-11}, 7.5 \times 10^{-9}]$ | $4.25 \times 10^{-10}$ | $[3.97 \times 10^{-10}, 4.53 \times 10^{-10}]$ | 6.48% |
| $D_{2,n}$  | Diffusion coefficient of electrolyte (Separator) | $m^2 \text{s}^{-1}$ | $[7.5 \times 10^{-11}, 7.5 \times 10^{-9}]$ | $8.61 \times 10^{-10}$ | $[7.02 \times 10^{-10}, 1.02 \times 10^{-9}]$ | 18.4% |
| $D_{2,n}$  | Diffusion coefficient of electrolyte (Negative) | $m^2 \text{s}^{-1}$ | $[7.5 \times 10^{-11}, 7.5 \times 10^{-9}]$ | $1.04 \times 10^{-9}$ | $[9.64 \times 10^{-10}, 1.12 \times 10^{-9}]$ | 7.64% |
| $\text{brugg}$  | Bruggeman's coefficient | — | — | 4* | — | — |
| $\theta(1)$  | Constant for conductivity equation of electrolyte | — | — | $[0.00413, 0.413]$ | $0.0381$ | $[0.0174, 0.0589]$ | 54.5% |
| $\theta(2)$  | Constant for conductivity equation of electrolyte | — | — | $[5 \times 10^{-4}, 5 \times 10^{-3}]$ | $5.45 \times 10^{-4}$ | $[5.30 \times 10^{-4}, 5.61 \times 10^{-4}]$ | 2.85% |
| $\theta(3)$  | Constant for conductivity equation of electrolyte | — | — | $[4.72 \times 10^{-8}, 4.72 \times 10^{-6}]$ | $4.79 \times 10^{-7}$ | $[4.69 \times 10^{-7}, 4.89 \times 10^{-7}]$ | 2.06% |
| $\theta(4)$  | Constant for conductivity equation of electrolyte | — | — | $[1.509 \times 10^{-11}, 1.509 \times 10^{-9}]$ | $1.48 \times 10^{-10}$ | $[1.43 \times 10^{-10}, 1.53 \times 10^{-10}]$ | 3.40% |
| $\theta(5)$  | Constant for conductivity equation of electrolyte | — | — | $[1.602 \times 10^{-15}, 1.602 \times 10^{-13}]$ | $1.46 \times 10^{-14}$ | $[1.35 \times 10^{-14}, 1.57 \times 10^{-14}]$ | 7.53% |
| $c_{1,p}$  | Initial solid phase concentration (Positive) | $\text{mol m}^{-3}$ | $[4236.5, 14236.5]$ | — | — | — |
| $c_{1,n}$  | Initial solid phase concentration (Negative) | $\text{mol m}^{-3}$ | — | 14300 | $[14300, 14300]$ | 0.0166% |
| $c_{2}$  | Initial concentration in the electrolyte | $\text{mol m}^{-3}$ | — | — | — | — |
| $E_a^p$  | Reaction constant activation energy (Positive) | $\text{J mol}^{-1}$ | — | 5000* | — | — |

(Continues)
| Symbol | Description | Unit | Input range [Min, Max] | Value | CI [Min, Max] | $R_{CI}$ (%) |
|--------|-------------|------|------------------------|-------|---------------|--------------|
| $E_{a}^{k}$ | Reaction constant activation energy (Negative) | J mol$^{-1}$ | — | 5000* | — | — |
| $E_{a}^{D_{1,p}}$ | Diffusion coefficient of solid phase activation energy (Positive) | J mol$^{-1}$ | — | 5000* | — | — |
| $E_{a}^{D_{1,n}}$ | Diffusion coefficient of solid phase activation energy (Negative) | J mol$^{-1}$ | — | 5000* | — | — |
| $E_{a}^{C_{p}}$ | Electrolyte conductivity activation energy (Positive) | J mol$^{-1}$ | — | 5000* | — | — |
| $E_{a}^{C_{n}}$ | Electrolyte conductivity activation energy (Separator) | J mol$^{-1}$ | — | 5000* | — | — |
| $E_{a}^{D_{1,p}}$ | Electrolyte conductivity activation energy (Negative) | J mol$^{-1}$ | — | 5000* | — | — |
| $E_{a}^{D_{2,p}}$ | Diffusion coefficient of electrolyte activation energy (Positive) | J mol$^{-1}$ | — | 5000* | — | — |
| $E_{a}^{D_{2,n}}$ | Diffusion coefficient of electrolyte activation energy (Separator) | J mol$^{-1}$ | — | 5000* | — | — |
| $E_{a}^{D_{1,n}}$ | Diffusion coefficient of solid phase activation energy (Negative) | J mol$^{-1}$ | — | 5000* | — | — |
| $l_{p}$ | Thickness (Positive) | m | $[8.0 \times 10^{-6}, 8.0 \times 10^{-4}]$ | $6.98 \times 10^{-3}$ | $[6.73 \times 10^{-3}, 7.23 \times 10^{-3}]$ | 13.0% |
| $l_{s}$ | Thickness (Separator) | m | $[2.5 \times 10^{-6}, 2.5 \times 10^{-4}]$ | $2.74 \times 10^{-5}$ | $[2.62 \times 10^{-5}, 2.86 \times 10^{-5}]$ | 4.39% |
| $l_{n}$ | Thickness (Negative) | m | $[8.8 \times 10^{-6}, 8.8 \times 10^{-4}]$ | $7.63 \times 10^{-4}$ | $[7.28 \times 10^{-4}, 7.98 \times 10^{-4}]$ | 4.54% |
| $A$ | Area of electrode | m$^{2}$ | [0.01, 1] | 0.09718 | [0.0932, 0.101] | 4.05% |
changes during operation and degradation over a cycle. The PIIA framework based on evaluating terminal voltages can increase the accuracy and robustness, including the electrochemical phenomena of a very short time scale. Therefore, we also will extend our studies to explore the voltage response of pulse currents that vary on short time scales under various operating conditions.

ACKNOWLEDGMENTS
This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korean government (Ministry of Science and ICT) (NRF-20151009350), Korea Institute of Energy Technology Evaluation and Planning (KETEP) grants funded by the Ministry of Trade, Industry & Energy, Republic of Korea (No.20214910100070), and Korea Institute of Science and Technology Information (KISTI) project (K-21-SG29-01R-1). This research was also supported by Seoul R&BD Program (SC210026) through the Seoul Business Agency (SBA) funded by The Seoul Metropolitan Government.

NOMENCLATURE
\( \alpha_a \) Cathodic charge transfer coefficient
\( \alpha_c \) Anodic charge transfer coefficient
\( \eta \) Overpotential, V
\( \text{FIM} \) Fisher information matrix
\( \phi \) Potential, V

\( \theta \) Constant for electrolyte conductivity
\( \epsilon \) Porosity
\( I_{\text{app}} \) Applied current density, A m\(^{-2}\)
\( R_p \) Particle radius, m
\( t^+ \) Transference number
\( U \) Open-circuit potential, V
\( V \) Voltage
\( \kappa \) Ionic conductivity, S m\(^{-1}\)
\( i \) Current density, A m\(^{-2}\)
\( \sigma \) Electronic conductivity, S m\(^{-1}\)
\( a \) Specific surface area, m\(^2\) m\(^{-3}\)
\( \text{brugg} \) Bruggeman’s coefficient
\( c \) Li+ concentration, mol m\(^{-3}\)
\( D \) Diffusivity, m\(^2\) s\(^{-1}\)
\( E \) Energy, J mol\(^{-1}\)
\( F \) Faraday’s constant, C mol\(^{-1}\)
\( j \) Pore wall flux of Li+, mol m\(^{-2}\) s\(^{-1}\)
\( k \) Reaction rate constant, m\(^{2.5}\) mol\(^{-0.5}\) s\(^{-1}\)
\( R \) Universal gas constant, J mol\(^{-1}\) K\(^{-1}\)
\( r \) Radial coordinate, m
\( T \) Temperature, K
\( t \) Time, s
\( x \) Spatial coordinate, m

Subscript
1 Solid-phase
2 Electrolyte-phase
obs Observation
ref Reference
k Domain indices
n Negative electrode
p Positive electrode
se Solid/Electrolyte interface
sn Interface between separator and negative electrode
sp Interface between separator and positive electrode

Superscript
avg Volume-averaged value
eff Effective value
max Maximum value

ORCID
Yun Young Choi https://orcid.org/0000-0002-6017-258X
Seongyoon Kim https://orcid.org/0000-0001-7326-5681
Kyunghyun Kim https://orcid.org/0000-0001-6527-1002
Sanghyun Kim https://orcid.org/0000-0003-1087-5727
Jung-Il Choi https://orcid.org/0000-0002-4948-1250

REFERENCES
1. Dunn B, Kamath H, Tarascon JM. Electrical energy storage for the grid: a battery of choices. Science. 2011;334(6058):928-935.
2. Oh KY, Samad NA, Kim Y, Siegel JB, Stefanopoulou AG, Epureanu BI. A novel phenomenological multi-physics model of Li-ion battery cells. J Power Sources. 2016;326:447-458.
3. Peng X, Chen S, Garg A, Rao N, Panda B. A review of the estimation and heating methods for lithium-ion batteries pack at the cold environment. *Energy Sci Eng*. 2019;7(3):645-662.

4. Huo Y, Hu W, Li Z, Rao Z. Research on parameter identification and state of charge estimation of improved equivalent circuit model of Li-ion battery based on temperature effects for battery thermal management. *Int J Energy Res*. 2020;44(14):11583-11596.

5. Shuai W, Li E, Wang H. An equivalent circuit model of a deformed Li-ion battery with parameter identification. *Int J Energy Res*. 2020;44(11):8372-8387.

6. Kim S, Choi YY, Choi JI. Impedance-based capacity estimation for lithium-ion batteries using generative adversarial network. *arXiv preprint arXiv:2107.06177* 2021.

7. Wang Y, Gao G, Li X, Chen Z. A fractional-order model-based state estimation approach for lithium-ion battery and ultra-capacitor hybrid power source system considering load trajectory. *J Power Sources*. 2020;449:227543.

8. Ruan H, He H, Wei Z, Quan Z, Li Y. State of health estimation of lithium-ion battery based on constant-voltage charging reconstruction. *IEEE J Emerg Sel Top Power Electron*. 2021;1–1.

9. Hu J, He H, Wei Z, Li Y. Disturbance-immune and aging-robust internal short circuit diagnostic for lithium-ion battery. *IEEE Trans Industr Electron*. 2021;69(2):1988–1999.

10. Wang S, Fernandez C, Fan Y, et al. A novel safety assurance method based on the compound equivalent modeling and iterate reduce particle-adaptive Kalman filtering for the unmanned aerial vehicle lithium ion batteries. *Energy Sci Eng*. 2020;8(5):1484-1500.

11. Cao W, Wang SL, Fernandez C, Zou CY, Yu CM, Li XX. A novel adaptive state of charge estimation method of full life cycling lithium-ion batteries based on the multiple parameter optimization. *Energy Sci Eng*. 2019;7(5):1544-1556.

12. Xu W, Xu J, Liu B, Liu J, Yan X. A multi-timescale adaptive dual particle filter for state of charge estimation of lithium-ion batteries considering temperature effect. *Energy Sci Eng*. 2020;8(8):2784-2798.

13. Li Y, Wei Z, Xiong B, Vilathgamuwa DM. Adaptive ensemble-based electrochemical-thermal-degradation state estimation of Lithium-Ion batteries. *IEEE Trans Industr Electron*. 2021;1–1.

14. Doyle M, Fuller TF, Newman J. Modeling of galvanostatic charge and discharge of the lithium/polymer/insertion cell. *J Electrochem Soc*. 1993;140(6):1526.

15. Hu X, Li S, Peng H. A comparative study of equivalent circuit models for Li-ion batteries. *J Power Sources*. 2012;198:359-367.

16. Feng F, Hu X, Hu L, Hu F, Li Y, Zhang L. Propagation mechanisms and diagnosis of parameter inconsistency within Li-ion battery packs. *Renew Sustain Energy Rev*. 2019;112:102-113.

17. Li J, Zou L, Tian F, Dong X, Zou Z, Yang H. Parameter identification of lithium-ion batteries model to predict discharge behaviors using heuristic algorithm. *J Electrochem Soc*. 2016;163(8):A1646.

18. Zhang L, Wang L, Hinds G, Lyu C, Zheng J, Li J. Multi-objective optimization of lithium-ion battery model using genetic algorithm approach. *J Power Sources*. 2014;270:367-378.

19. Forman JC, Moura SI, Stein JL, Fathy HK. Genetic identification and fisher identifiability analysis of the Doyle-Fuller-Newman model from experimental cycling of a LiFePO4 cell. *J Power Sources*. 2012;210:263-275.

20. Deng Z, Deng H, Yang L, Cai Y, Zhao X. Implementation of reduced-order physics-based model and multi-parameters identification strategy for lithium-ion battery. *Energy*. 2017;138:509-519.

21. Pang H, Mou L, Guo L, Zhang F. Parameter identification and systematic validation of an enhanced single-particle model with aging degradation physics for Li-ion batteries. *Electrochim Acta*. 2019;307:474-487.

22. Kim M, Chun H, Kim J, et al. Data-efficient parameter identification of electrochemical lithium-ion battery model using deep Bayesian harmony search. *Appl Energy*. 2019;254:113644.

23. Rahman MA, Anwar S, Izadain A. Electrochemical model parameter identification of a lithium-ion battery using particle swarm optimization method. *J Power Sources*. 2016;307:86-97.

24. Liu Y, Tang S, Li L, et al. Simulation and parameter identification based on electrochemical-thermal coupling model of power lithium-ion battery. *J Alloy Compd*. 2020;844:156003.

25. El-Sehiemy RA, Hamida M, Mesbah T. Parameter identification and state-of-charge estimation for lithium-polymer battery cells using enhanced sunflower optimization algorithm. *Int J Hydrogen Energy*. 2020;45(15):8833-8842.

26. Ferahtia S, Djerou A, Rezk H, Chouder A, Houari A, Machmoum M. Optimal parameter identification strategy applied to lithium-ion battery model. *Int J Energy Res*. 2021;45:16741-16753.

27. Chu Z, Jobman R, Rodriguez A, et al. A control-oriented electrochemical model for lithium-ion battery. Part II: Parameter identification based on reference electrode. *J Energy Storage*. 2020;27:101101.

28. Fan G. Systematic parameter identification of a control-oriented electrochemical battery model and its application for state of charge estimation at various operating conditions. *J Power Sources*. 2020;470:228153.

29. Ramadesigan V, Chen K, Burns NA, Boovaragavan V, Braatz RD, Subramaniam VR. Parameter estimation and capacity fade analysis of lithium-ion batteries using reformulated models. *J Electrochem Soc*. 2011;158(9):A1048.

30. Sitterly M, Yin GG, Wang C, et al. Enhanced identification of battery models for real-time battery management. *IEEE Trans Sustain Energy*. 2011;2(3):300-308.

31. Rothenberger MJ, Docimo DJ, Ghanaatpishe M, Fathy HK. Genetic optimization and experimental validation of a test cycle that maximizes parameter identifiability for a Li-ion equivalent-circuit battery model. *J Energy Storage*. 2015;4:156-166.

32. Song Z, Hofmann H, Lin X, Han X, Hou J. Parameter identification of lithium-ion battery pack for different applications based on Cramer-Rao bound analysis and experimental study. *Appl Energy*. 2018;231:1307-1318.

33. Park S, Kato D, Gima Z, Klein R, Moura S. Optimal experimental design for parameterization of an electrochemical Lithium-ion battery model. *J Electrochem Soc*. 2018;165(7):A1309.

34. Pozzi A, Ciaramella G, Volkwein S, Raimondo DM. Optimal design of experiments for a lithium-ion cell: parameters identification of an isothermal single particle model with electrolyte dynamics. *Ind Eng Chem Res*. 2018;58(3):1286-1299.

35. Lai Q, Jangra S, Ahn HJ, Kim G, Joe WT, Lin X. Analytical derivation and analysis of parameter sensitivity for battery electrochemical dynamics. *J Power Sources*. 2020;472:228338.

36. Vazquez-Arenas J, Gimenez LE, Fowler M, Han T, Sk C. A rapid estimation and sensitivity analysis of parameters describing
the behavior of commercial Li-ion batteries including thermal analysis. *Energy Convers Manage*. 2014;87:472-482.

37. Jin N, Danilov DL, van den Hof PMJ, Donkers M. Parameter estimation of an electrochemistry-based lithium-ion battery model using a two-step procedure and a parameter sensitivity analysis. *Int J Energy Res*. 2018;42(7):2417-2430.

38. Choi YY, Kim S, Kim S, Choi H. Multiple parameter identification using genetic algorithm in vanadium redox flow batteries. *J Power Sources*. 2020;450:227684.

39. Kim S, Choi YY, Kim KJ, Choi JI. Forecasting state-of-health of lithium-ion batteries using variational long short-term memory with transfer learning. *J Energy Storage*. 2021;41:102893.

40. Ramadesigan V, Boovaragavan V, Pirkle JC Jr, Subramanian VR. Efficient reformulation of solid-phase diffusion in physics-based lithium-ion battery models. *J Electrochem Soc*. 2010;157(7):A854.

41. Brun R, Reichert P, Kiinsch HR. Practical identifiability analysis of large environmental simulation models. *Water Resour Res*. 2001;37(4):1015-1030.

42. Franceschini G, Macchietto S. Model-based design of experiments for parameter precision: state of the art. *Chem Eng Sci*. 2008;63(19):4846-4872.

43. Feng A, Boraey M, Gwin M, Finlay P, Kuehl P, Vehring R. Mechanistic models facilitate efficient development of leucine containing microparticles for pulmonary drug delivery. *Int J Pharm*. 2011;409(1-2):156-163.

44. Machado VC, Tapia G, Gabriel D, Lafuente J, Baeza JA. Systematic identifiability study based on the Fisher Information Matrix for reducing the number of parameters calibration of an activated sludge model. *Environ Model Softw*. 2009;24(11):1274-1284.

45. Schmidt AP, Bitzer M, Aw I, Guzzella L. Experiment-driven electrochemical modeling and systematic parameterization for a lithium-ion battery cell. *J Power Sources*. 2010;195(15):5071-5080.

46. Mitchell M. *An Introduction to Genetic Algorithms*. MIT Press; 1998.

47. Kim S, Lee S, Choi JI, Cho H. Binary genetic algorithm for optimal jointpoint detection: application to cancer trend analysis. *Stat Med*. 2021;40(3):799-822.

48. Helton JC, Davis FJ. Latin hypercube sampling and the propagation of uncertainty in analyses of complex systems. *Reliab Eng Syst Saf*. 2003;81(1):23-69.

49. Torchio M, Magni L, Gopalanui RB, Braatz RD, Raimondo DM. Lionsimba: a MATLAB framework based on a finite volume model suitable for Li-Ion battery design, simulation, and control. *J Electrochem Soc*. 2016;163(7):A1192-A1205.

50. Northrop PW, Ramadesigan V, De S, Subramanian VR. Coordinate transformation, orthogonal collocation, model reformulation and simulation of electrochemical-thermal behavior of Lithium-Ion battery stacks. *J Electrochem Soc*. 2011;158(12):A1461.

51. Subramanian VR, Boovaragavan V, Ramadesigan V, Arabandi M. Mathematical model reformulation for Lithium-Ion battery simulations: Galvanostatic boundary conditions. *J Electrochem Soc*. 2009;156(4):A260.

52. Santhanagopalan S, Guo Q, Ramadass P, White RE. Review of models for predicting the cycling performance of Lithium Ion batteries. *J Power Sources*. 2006;156(2):620-628.

53. Laue V, Roder F, Krewer U. Practical identifiability of electrochemical P2D models for Lithium-Ion batteries. *J Appl Electrochem*. 2021;51:1253-1265.

54. He W, Williard N, Osterman M, Pecht M. Prognostics of Lithium-Ion batteries based on Dempster—Shafer theory and the Bayesian Monte Carlo method. *J Power Sources*. 2011;196(23):10314-10321.

55. Kim S, Choi Y Y, Choi J-II. Impedance-based capacity estimation for lithium-ion batteries using generative adversarial network. *Applied Energy*. 2022;308:118317.

**How to cite this article:** Choi YY, Kim S, Kim K, Kim S, Choi J-I. Parameter identification and identifiability analysis of lithium-ion batteries. *Energy Sci Eng*. 2022;10:488–506. doi: [10.1002/ese3.1039](https://doi.org/10.1002/ese3.1039)