Extending Life of Lithium-Ion Battery Systems by Embracing Heterogeneities via an Optimal Control-Based Active Balancing Strategy

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Abstract—This article formulates and solves a multiobjective fast charging-minimum degradation optimal control problem (OCP) for a lithium-ion battery module made of series-connected cells equipped with an active balancing circuitry. The cells in the module are subject to heterogeneity induced by manufacturing defects and nonuniform operating conditions. Each cell is expressed via a coupled nonlinear electrochemical, thermal, and aging model and the direct collocation approach is employed to transcribe the OCP into a nonlinear programming problem (NLP). The proposed OCP is formulated under two different schemes of charging operation: 1) same charging time (OCP-SCT) and 2) different charging time (OCP-DCT). The former assumes simultaneous charging of all cells irrespective of their initial conditions, whereas the latter allows for different charging times of the cells to account for heterogeneous initial conditions. The problem is solved for a module with two series-connected cells with intrinsic heterogeneity among them in terms of state of charge and state of health. Results show that the OCP-DCT scheme provides more flexibility to deal with heterogeneity, boasting of lower temperature increase, charging current amplitudes, and degradation. Finally, a comparison with the common practice of constant current (CC) charging over a long-term cycling operation shows that promising savings, in terms of retained capacity, are attainable under both control (OCP-SCT and OCP-DCT) schemes.

Index Terms—Aging, batteries, electrochemical modeling, optimal control.

NOMENCLATURE

- $c_{s,i}$ Concentration in solid phase [mol/m³].
- c_e Concentration in electrolyte phase [mol/m³].
- c_{solv} Solvent concentration [mol/m³].
- T_c Cell core temperature [K].
- $T_{\rm s}$ Cell surface temperature [K].
- *L*_{sei} SEI layer thickness [m].
- Q Cell capacity [Ah].
- I_{cell} Cell current [A].
- η_j Overpotential [V].

Manuscript received 1 March 2022; revised 4 August 2022; accepted 14 October 2022. Date of publication 3 November 2022; date of current version 25 April 2023. This work was supported by LG Chem (now LG ES). Recommended by Associate Editor D. M. Raimondo. (*Corresponding author: Simona Onori.*)

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Color versions of one or more figures in this article are available at https://doi.org/10.1109/TCST.2022.3215610.

Digital Object Identifier 10.1109/TCST.2022.3215610

Exchange current density [A/m²]. $i_{0, i}$ U_i Open-circuit potential (electrode) [V]. $V_{\rm oc}$ Open-circuit voltage (cell) [V]. i_s Side-reaction current density [A/m³]. $D_{s,i}$ Solid-phase diffusion $[m^2/s]$. $R_{s,j}$ Particle radius [m]. Specific interfacial surface area $[m^{-1}]$. $a_{s,j}$ Α Cell cross-sectional area $[m^2]$. L_i Domain thickness [m]. F Faraday's constant [C/mol]. c^{\max} Maximum electrode concentration [mol/m³]. κ_j^{eff} Effective electrolyte conductivity [S/m]. k_{j} Reaction rate constant [m^{2.5}/s-mol^{0.5}]. R_l Lumped contact resistance $[\Omega]$. $R_{\rm el}$ Electrolyte resistance $[\Omega]$. R_{sei} SEI layer resistance $[\Omega]$. R_{g} Universal gas constant [J/mol-K]. Solvent diffusion in SEI layer $[m^2/s]$. D_{solv} SEI layer porosity. $\epsilon_{\rm sei}$ SEI layer density [kg/m³]. $\rho_{\rm sei}$ SEI layer ionic conductivity [S/m]. κ_{sei} Solvent concentration [mol/m³]. c_{solv} $M_{\rm sei}$ Molar mass of SEI layer [kg/mol]. β Side reaction charge transfer coefficient. C_s Heat capacity of cell surface [J/K]. C_c Heat capacity of cell core [J/K]. R_c Conductive resistance—core/surface [K/W]. R_u Convective resistance-surface/surroundings [K/W]. T_{amb} Ambient temperature [K]. $N_{r,i}$ Number of radial discretization points. N_{sei} Number of SEI layer discretization points. $c_{s,j}^{\text{surf}}$ c_{solv}^{surf} c_{e}^{surf} θ_{i}^{surf} Surface concentration in solid phase [mol/m³]. Surface solvent concentration [mol/m³]. Average electrolyte concentration [mol/m³]. surface stoichiometry in solid phase. $b_j \\ c_{s,j}^{\text{bulk}}$ Bulk concentration [mol/m³]. $E_{a,\varphi}$ Activation energy [J/mol]. $\theta_{0\%}^{J}$ Reference stoichiometry ratio at 0% SOC. $\theta_{100\%}^{j}$ Reference stoichiometry ratio at 100% SOC. k_f Solvent reduction rate constant $[mol^{-2}s^{-1}]$. Optimal solvent concentration [mol/m³]. c_{solv}^* SEI layer ionic conductivity [S/m]. κ_{sei}

 I_{cell} Cell current [A].

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V_{cell}	Cell voltage [V].		
$T_{c,\mathrm{ref}}$	Reference core temperature [K].		
$\epsilon_{e,j}$	Electrolyte porosity.		
$\Phi_{s,n}$	Anode surface potential [V].		
U_s	Solvent reduction potential [V].		
r	Radial coordinate.		
R_m	Cell-to-cell heat transfer resistance [K/W]		
N _{cell}	Number of cells.		
x	State vector.		
I_0	Module current [A].		
I_B	Balancing current [A].		
X	Optimization variable vector.		
t _f	Charging time [s].		
N_s	Number of states.		
$N_{\rm BP}$	Number of break points.		
$Q_{ m nom}$	Nominal capacity [Ah].		
$\alpha, \beta_1, \beta_2, \beta_3$	Optimization parameters.		
S _p	Smoothness degree.		
d_p	Polynomial order.		
P	Free parameter set.		
μ_1, μ_2	Karush–Kuhn–Tucker multipliers.		
$\mathcal{B}_{p,q}$	B-Spline.		
$\omega_{p,q}$	Free parameters of optimization variables		
ϕ	Thermal diffusivity.		
R _{cell}	Radius of a cylindrical 18650 cell [m].		
BP	Break point.		
CCCV	Constant-current constant-voltage.		
СР	Collocation point.		
DAE	Differential algebraic equation.		
DOD	Depth of discharge.		
DCT	Different charging time.		
FDM	Finite difference method.		
GQF	Gaussian quadrature formula.		
IPOPT	Interior point optimizer.		
KKT	Karush–Kuhn–Tucker.		
LIB	Lithium-ion battery.		
NLP	Nonlinear programming.		
NMC	Nickel-manganese-cobalt.		
OCP	Optimal control problem.		
ODE	Ordinary differential equation.		
PDE	Partial differential equation.		
SCT	Same charging time.		
SEI	Solid electrolyte interphase.		
SOC	State of charge.		
SOH	State of health.		
SPM	Single particle model.		

I. INTRODUCTION

THE LIBs are the enabling technology to ensure a sustainable future due to their high cell voltage, high energy and power density, low memory effect, long life, and increasingly reduced cost [1]. They have been extensively utilized in a wide range of applications, including microgrids, consumer electronics, and electric vehicles (EVs) [2], [3], [4]. Consumer acceptance of battery-powered devices is highly dependent on their fast charging ability while maintaining a safe and long-running operation. In EVs today, constant current (CC) charging is used, where the charger supplies a relatively uniform current, regardless of the battery SOC or temperature [5], [6]. Batteries used in EVs consist of a large number of cells connected both in series and parallel. Variations in the parameters of individual battery cells, such as capacity mismatch, impedance, and operating temperature, are deemed to expand throughout the life of the device. One of the tasks of the battery management system (BMS) is to provide cell balancing functionality by measuring and comparing the states of all cells after each charging cycle. Recent advances in battery life management have come from advanced BMS strategies that rely on battery models around which estimation and optimization strategies are designed. Much of the recent battery control/optimization literature, though, has focused on single-cell operation under fast charging. Methods to optimizing longevity under fast charging operation for single cells have been proposed based on model predictive control (MPC) [7], [8], NLP [9], [10], and control vector parameterization (CVP) [11] either using equivalent circuit models or electrochemical models. However, the problem of battery fast charging while preserving its health is a pack-level challenge that needs to be tackled as such.

A. Motivation and Related Literature

A battery pack consists of individual cells, which are organized into modules made of cells connected in series/parallel. Results obtained for single cells cannot be extrapolated or generalized to the module/pack level due to the loss of modularity in the system [12]. A fundamental characteristic of an interconnected battery system (module and pack) is that heterogeneity in the parameters within the series/parallel cells is inevitable due to manufacturing and operating conditions, which, if not monitored or corrected on time, could hinder the performance and longevity of the battery system during operation [13]. Manufacturing-induced heterogeneities, such as capacity and impedance of single cells, are deemed to be exacerbated over time and, at the same time, end up being the cause of differences in temperature, SOC, DOD, and charging rate [14], [15], [16]. For example, voltage and charge imbalances limit the charge/discharge capabilities of the pack, posing limitations on pack-level performance and causing temperature imbalance, which is known to accelerate battery pack aging [17].

Battery equalization methods are employed to bring the cells in a pack to the same voltage/SOC [18]. These methods fall into two main categories: passive and active balancing. In passive methods, for example, in the form of a fixed shunting resistor, no active control is used to balance the cells and the excess energy from the high SOC cells is dissipated until the charge matches the lower SOC cells in the pack. Active balancing methods, on the other hand, offer more flexibility in equalizing the energy of each cell in the pack [18] and rely on active control strategies. It is worth mentioning that in the literature, there is a lack of consensus as to what is interpreted as an active or passive balancing framework. In some cases, energy storing and redistributing components,

such as dc/dc converters, are considered to constitute an active balancing circuit, and in other cases, the presence of a control strategy to balance the cells (either through switching shunt resistors, transistors, or dc/dc converters) is considered to constitute an active balancing circuit. For example, in cell-bypass active methods, implemented either via shunt resistor or shunt transistor method [18], the current of each cell is bypassed whenever the cell voltage reaches the admissible upper limit by means of a switch in series with a resistor or a transistor, respectively. In the cell-to-cell methods, in the form of, for example, bypass dc/dc converters [19], the extra energy stored in the most charged cells is transferred to the least charged cells. Alternatively, balancing and complete cell-bypassing can be achieved by a module-integrated distributed battery system architecture [20], in which each cell in the module is individually managed by the modular converter without the need for equalization circuits. The proposed work falls in the category of active balancing, in accordance with [18], since an optimal controller is proposed to actively switch the shunt resistors in cell-bypass balancing methods or switch dc/dc converters in cell-to-cell balancing methods.

While hardware strategies to enable active balancing are in place, scant attention has been paid to synthesizing optimization-based control strategies for battery pack/module. The impact of different balancing strategies on cell-to-cell variations, in terms of SOC, maximum capacity, and resistance, is addressed in [21], where a formal framework based on linearized electrochemical dynamics and multivariable control theory is used to: 1) show that voltage balancing fails to eliminate capacity and resistance imbalance between cells and 2) design a strategy that is able to eliminate charge, capacity, and resistance imbalance within the lifespan of the pack. In [22], an electrothermal control scheme is devised for load management of a battery module for onboard vehicle operation to tackle charge and temperature imbalances by leveraging constrained linear quadratic MPC. In [23], charge imbalance and temperature imbalance are also tackled by using a formal framework based on MPC to obtain insights on how temperature imbalance can be controlled through an average current. A simplified linear parameter varying model is developed to represent charge and temperature imbalance. In [24], SOC imbalance in series-connected cells is controlled via a nonlinear MPC scheme upon proper simplifications of the electrochemical battery dynamics and insights on an easily implementable power supply scheme are provided.

B. Main Contributions

In this article, the system under investigation is an LIB module of N_{cell} series-connected cells (see Fig. 1), where each cell is connected to active balancing hardware, which could be either as simple as an active shunt resistor or shunt transistor method or more sophisticated hardware such as bypass dc/dc converters.¹ For the given system, we address the problem of designing an optimization-based control strategy that controls individual cells to achieve fast charging while guaranteeing

minimum degradation of the pack to be implemented in active balancing hardware. In Fig. 1, the current of the *k*th cell is given by $I_{cell_k} = I_0 - I_{B_k}$, where I_0 is the module current and I_{B_k} is the current absorbed by the balancing hardware associated with *k*th cell. Battery pack life optimization is achieved by controlling each individual cell while embracing heterogeneities in terms of state and parameters—due to either/both manufacturing defects or/and nonuniform operating conditions. The formulated OCP will ultimately implement SOC balancing along with SOH-aware balancing by tackling the cell-to-cell heterogeneity. The optimal control is multiobjective in nature to face the conflicting objectives of minimum time of charge (t_{f_k}) under minimum degradation by optimizing the current profiles.²

Cells in the module are modeled via coupled nonlinear PDEs, ODEs, and DAEs describing the electrochemical, thermal, and aging dynamics [25]. The SPM is employed to model the electrochemical dynamics, a lumped two-state thermal model with cell-to-cell heat transfer terms is used to derive the core-cell temperature from ambient temperature, and finally, aging is modeled through the growth of SEI layer on the negative electrode.

Within the framework adopted, the cell is a multi-time-scale system in which thermal dynamics acts as fast dynamics, the electrochemical dynamics as semislow dynamics, and aging dynamics as slow dynamics [17]. The nonlinear and multitime-scale nature of the cell dynamics are retained in the formulation and solution of the multiobjective optimization problem addressed in this work, and to the best of our knowledge, to date, no study has addressed such a problem using the high-fidelity multi-time-scale battery model. Note the fact that aging dynamics includes SEI layer growth and solvent concentration, where the former is a low-dimensional slow variable and the latter is a high-dimensional one. The computational burden imposed by the high-fidelity dynamics at different time scales has led to the design of a surrogate model to approximate the high-dimensional slow dynamics (solvent concentrations) as a function of cell current and ambient temperature.

To solve the optimization problem, the direct collocation approach [26] is utilized to transcribe the OCP to an NLP [27] by parameterization of the system states and inputs, and charging times. The interior point solver IPOPT [28] is then used to solve the NLP problem, while the optimality of the solution is discussed using the KKT conditions (first-order necessary conditions). The OCP is formulated under two different schemes: 1) SCT (OCP-SCT) and 2) DCT (OCP-DCT). To confirm the soundness of the proposed OCP-SCT and OCP-DCT schemes, simulation studies are carried out on an illustrative example of a battery module with two series-connected cells, each equipped with active balancing hardware. The performance and robustness of the proposed schemes are shown under perturbation of parameters in terms of initial SOC and initial SOH imbalances (through variation in the initial SEI layer growth state).

¹The specific hardware design is outside the scope of this article. The reader can refer to [18] for different active hardware balancing solutions.

²High C-rate currents would charge the battery faster at the expense of faster growth of SEI layer, causing capacity and power fade.



Fig. 1. Battery module with N_{cell} series-connected cells, where each cell is connected to balancing hardware. The current variable I_0 refers to the module current, variables $I_{cell_1}, \ldots, I_{cell_{N_{cell}}}$ are the current magnitude through the cells, variables $I_{B_1}, \ldots, I_{B_{N_{cell}}}$ are the balancing currents, and time variables $t_{f_1}, \ldots, t_{f_{N_{cell}}}$ are the charging time associated with each cell.

This article extends on the initial investigation proposed in [29] in that: 1) it contains the description of the surrogate model used to capture the solvent diffusion dynamics of the SEI layer growth model; 2) it characterizes the time-scale difference of the LIB dynamics; 3) it provides ample simulation scenarios of the two optimization schemes for an effective and exhaustive comparison of the two; and 4) it provides a comparative study of the two charging scenarios with the traditionally used CC charging protocol. The main takeaways and recommendations from the proposed study are provided in the pursuit of a novel life-extension optimization charging strategy that embraces cell-to-cell heterogeneities by combining advanced optimization algorithms over multiscale high-fidelity models using active balancing hardware setup.

C. Outline

The organization of this article is given as follows. Section II lists the notations used in this article. Section III presents the mathematical model for cells and battery modules. Section IV describes the problem statement. Section V formulates the proposed optimal control methodology. Section VI presents the simulation results. Section VII presents the discussion and conclusion.

II. NOTATIONS

The following notations are used in this article.

- Given a real *n*-dimensional vector x with initial and final values x(t₀) and x(t_f) (t₀ and t_f are the initial and final times), Δx = ((|x(t_f) x(t₀)|)/(x(t₀))) × 100% is the percentage deviation of x with respect to its initial value.
- 2) Given the continuously differentiable function f(x), $\nabla f(x)$ is the gradient of f(x) with respect to x.
- 3) The subscript $j \in [n; p]$ stands for the cell domain (e.g. n = anode and p = cathode).
- 4) The subscript *i* refers to the discretization grid position when converting PDEs to ODEs via FDM in solid electrodes and SEI layer spatial dimensions.
- 5) The superscript k represents the cell position within the series-connected module.

III. BATTERY MODULE MODEL

This section presents the model of the LIB module with N_{cell} series-connected cells. Each cell is equipped with an active

balancing circuitry that provides a practical way to reroute the current flowing in each cell and that is used as an extra degree of freedom to the optimal controller. The variables used in this article are listed in the Nomenclature.

A. Cell Electrochemical Model

The SPM used to model the cell electrochemical dynamics assumes that each electrode is a single spherical particle and that the concentration gradient in the electrolyte phase is uniform; hence, the diffusion electrolyte dynamics can be neglected. SPM is described by two governing PDEs—one for each electrode—representing the mass conservation in the solid phase through Fick's law

$$\frac{\partial c_{s,j}}{\partial t} = \frac{D_{s,j}(T_c)}{r^2} \frac{\partial}{\partial r} \left[r^2 \frac{\partial c_{s,j}}{\partial r} \right], \qquad j \in [n, p]$$
(1)

associated with the Neumann boundary conditions at the center and surface of the spherical particle given by

$$\frac{\partial c_{s,j}}{\partial r}\Big|_{r=0} = 0$$

$$\frac{\partial c_{s,j}}{\partial r}\Big|_{r=R_{s,j}} = \frac{\pm I_{\text{cell}}}{D_{s,j}(T)a_{s,j}AL_jF} + g_{s,j}$$
(2)

where $g_{s,j}$ is a nonlinear function of $c_{s,j}^{\text{surf}}, c_{\text{solv}}^{\text{surf}}, T_c, I_{\text{cell}}$, and L_{sei} . At the boundary of the particle when r = $R_{s,i}$, the right-hand side (RHS) of the boundary condition in (2) has a negative sign for the negative electrode, whereas the positive sign is for the positive electrode. The sign is considered to indicate the intercalation and de-intercalation of lithium within the positive and negative electrodes. For instance, when the cell is being discharged $(I_{cell} > 0)$, the RHS sign $((\partial c_{s,n}/\partial r)|_{r=R_{s,n}} < 0)$ indicates that lithium is being de-intercalated at the negative electrode (due to the oxidation reaction) and intercalated $((\partial c_{s,p}/\partial r)|_{r=R_{s,p}} > 0)$ at the positive electrode (due to the reduction reaction). During charging ($I_{cell} < 0$), the RHS sign indicates intercalation $((\partial c_{s,n}/\partial r)|_{r=R_{s,n}} > 0)$ at the negative electrode and de-intercalation at the positive electrode $((\partial c_{s,p}/\partial r)|_{r=R_{s,p}} < 0)$. The complete expression of the function $g_{s,j}$ for each electrode is reported in (18). We use the FDM to radially discretize the PDEs (1) into a system of ODEs [25]. Solid electrode parameters, including the diffusion coefficient $D_{s,i}$ and the reaction rate constant k_i , follow an

Arrhenius relationship with temperature given by:

$$\varphi(T_c) = \varphi_{\text{ref}} \exp\left[\frac{E_{a,\varphi}}{R_g} \left(\frac{1}{T_{c,\text{ref}}} - \frac{1}{T_c}\right)\right]$$
(3)

where $T_{c,ref} = 298$ K, φ to be either $D_{s,j}$, D_{solv} , or k_j , and φ_{ref} is the value of φ at reference temperature $T_{c,ref}$.

The surface overpotentials of each electrode, η_j for $j \in [n, p]$, are obtained from the Butler–Volmer kinetic equation describing the rate of intercalation and de-intercalation of lithium ions as

$$\eta_j = \frac{R_g T_c}{0.5F} \sinh^{-1} \left(\frac{I_{\text{cell}}}{2Aa_{s,j} L_j i_{0,j}} \right), \quad j \in [n, p]$$
(4)

where the exchange current density $i_{0,j}$ is given by

$$i_{0,j} = k_j F_{\sqrt{c_e^{\text{avg}} c_{s,j}^{\text{surf}} \left(c_{s,j}^{\text{max}} - c_{s,j}^{\text{surf}} \right)}, \quad j \in [n, p].$$
 (5)

The cell voltage V_{cell} can be calculated as

$$V_{\text{cell}} = U_p(c_{s,p}^{\text{surf}}) + \eta_p(c_{s,p}^{\text{surf}}, c_e^{\text{avg}}, T_c, I_{\text{cell}}) - U_n(c_{s,n}^{\text{surf}}) - \eta_n(c_{s,n}^{\text{surf}}, c_e^{\text{avg}}, T_c, I_{\text{cell}}) - I_{\text{cell}}(R_l + R_{\text{el}} + R_{\text{sei}})$$
(6)

in which the cell ohmic resistance includes the contact resistance R_l , electrolyte resistance R_{el} , and SEI layer resistance R_{sei} , where the last two parameters are expressed as

$$R_{\rm el} = \frac{1}{2A} \left[\frac{L_n}{\kappa_{e,n}^{\rm eff}} + \frac{2L_s}{\kappa_{e,s}^{\rm eff}} + \frac{L_p}{\kappa_{e,p}^{\rm eff}} \right]$$
$$R_{\rm sei} = \frac{L_{\rm sei}}{a_{s,n}AL_n\kappa_{\rm sei}}$$
(7)

where $\kappa_{e,j}^{\text{eff}}$ is a function of c_e^{avg} , and $\epsilon_{e,n}$, $\epsilon_{e,s}$, and $\epsilon_{e,p}$ are the porosity values in the negative electrode, separator, and positive electrode, respectively. The cell voltage is also dependent on the open-circuit potentials of electrodes U_j , with $j \in [n, p]$, that are calculated using empirical relationships as functions of electrode surface concentration stoichiometry [17], [25] (also shown in Fig. 5 for the cell chemistry used in this study).

The bulk SOC of each electrode is given by

$$SOC_{j}^{bulk} = \frac{\frac{c_{s,j}^{c_{s,j}}}{c_{s,j}^{max}} - \theta_{0\%}^{j}}{\theta_{100\%}^{j} - \theta_{0\%}^{j}}, \quad j \in [n, p]$$
(8)

that varies between two stoichiometric values $\theta_{100\%}^{j}$ and $\theta_{0\%}^{j}$, representing fully charged and discharged conditions for each electrode. In this article, SOC^{bulk} is used as battery cell SOC in the optimization algorithm as the cathode is the limiting electrode.

B. Cell Thermal Model

The thermal dynamics is modeled using the lumped parameter two-state thermal model

$$C_c \frac{dT_c}{dt} = I_{\text{cell}}(V_{\text{oc}} - V_{\text{cell}}) + \frac{T_s - T_c}{R_c}$$

$$C_s \frac{dT_s}{dt} = \frac{T_{\text{amb}} - T_s}{R_u} - \frac{T_s - T_c}{R_c}$$
(9)

where T_c and T_s are the core and surface temperature of each cell, respectively. This model assumes that the internal temperature is uniformly distributed across the core and the surface temperature is uniform throughout the surface [30].

C. Cell Aging Model

A physics-based approach is employed for battery aging that considers the anode SEI layer growth as a function of solvent reduction kinetics and diffusion dynamics to predict cell capacity loss and power fade. For the radial coordinate $r \in [R_{s,n}, R_{s,n} + L_{sei}]$ across the thickness of the SEI layer, the solvent concentration available for reduction reaction at the anode surface is modeled by

$$\frac{\partial c_{\text{solv}}}{\partial t} = D_{\text{solv}}(T_c) \frac{\partial^2 c_{\text{solv}}}{\partial r^2} - \frac{dL_{\text{sei}}}{dt} \frac{\partial c_{\text{solv}}}{\partial r}$$
(10)

with boundary conditions

$$-D_{\text{solv}}(T_c)\frac{\partial c_{\text{solv}}}{\partial r}\Big|_{r=R_{s,n}} + \frac{dL_{\text{sei}}}{dt}c_{\text{solv}}^{\text{surf}} = \frac{i_s}{F}$$
$$c_{\text{solv}}\Big|_{r=R_{s,n}+L_{\text{sei}}} = \epsilon_{\text{sei}}c_{\text{solv}}^{\text{bulk}}.$$
 (11)

The PDE aging dynamics (10) is discretized via FDM where a time-varying grid size is used to account for changes in the SEI layer thickness [25]. The SEI layer growth is modeled as follows:

$$\frac{dL_{\rm sei}}{dt} = -\frac{i_s M_{\rm sei}}{2F\rho_{\rm sei}} \tag{12}$$

where the rate of change of L_{sei} is linearly proportional to the side-reaction current

$$i_{s} = -2Fk_{f} \left(c_{s,n}^{\text{surf}}\right)^{2} c_{\text{solv}}^{\text{surf}} \\ \times \exp\left[\frac{-\beta F}{R_{g}T_{c}} \left(\Phi_{s,n} - R_{\text{sei}}I_{\text{cell}} - U_{s}\right)\right].$$
(13)

The capacity loss is modeled by integrating the side-reaction current as

$$\frac{dQ}{dt} = i_s A L_n a_{s,n}.$$
 (14)

D. State-Space Representation: Cell Level

Upon discretization, the governing PDEs are transformed into a system of ODEs and DAEs using which the celllevel state-space form can be derived. Note that DAEs are related to the Butler–Volmer equation used to calculate the overpotentials.

1) Solid-Phase Diffusion: The state-space representation of the solid-phase diffusion dynamics for each electrode is represented as

$$\dot{\boldsymbol{c}}_{s,j} = \alpha_{s,j} A_{s,j} \boldsymbol{c}_{s,j} + \beta_{s,j} B_{s,j} [I_{\text{cell}} - g_{s,j}]$$
(15)

where $c_{s,j} = [c_{s,j,1}, \dots, c_{s,j,N_{r,j}}]^T \in \mathbb{R}^{N_{r,j}}$ with $c_{s,j,N_{r,j}} = c_{s,j}^{\text{surf}}$, $B_{s,j} = [0, \dots, (2 + (2/(N_r - 1)))]^T \in \mathbb{R}^{N_{r,j}}$

$$A_{s,j} = \begin{bmatrix} -2 & 2 & 0 & 0 & \cdots & 0 & 0 \\ 1/2 & -2 & 3/2 & 0 & \cdots & 0 & 0 \\ 0 & 2/3 & -2 & 4/3 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \cdots & 2 & -2 \end{bmatrix} \in \mathbb{R}^{N_{r,j} \times N_{r,j}}$$
(16)

$$\alpha_{s,j} = \frac{D_{s,j}(T_c)}{\Delta r_j^2}, \quad \beta_{s,j} = \begin{cases} \frac{-1}{AL_j F a_{s,j} \Delta r_j}, & \text{if } j = n\\ \frac{1}{AL_j F a_{s,j} \Delta r_j}, & \text{if } j = p \end{cases}$$
(17)

and

$$g_{s,j}(c_{s,j}^{\text{surf}}, c_{\text{solv}}^{\text{surf}}, T_c, I_{\text{cell}}, L_{\text{sei}}) = \begin{cases} a_{s,n}L_nAi_s, & \text{if } j = n\\ 0, & \text{if } j = p \end{cases}$$
(18)

with $\Delta r_j = (R_{s,j}/(N_{r,j}-1))$ and $N_{r,j}$ the number of radial discretization grids in SPM.

2) SEI Layer Growth: The ODEs for SEI layer growth and capacity loss are given by

$$\dot{L}_{sei} = \beta_{sei} g_{s,n}$$
 and $\dot{Q} = \frac{\dot{L}_{sei}}{\beta_{sei}} = a_{s,n} L_n A i_s$ (19)

with $\beta_{\text{sei}} = (-M_{\text{sei}}/(2F\rho_{\text{sei}}a_{s,n}L_nA)).$

3) Solvent Diffusion: The ODEs describing the solvent diffusion dynamics are given by

$$\dot{c}_{\text{solv}} = \begin{cases} 2\alpha_{\text{solv}}(c_{\text{solv},2} - c_{\text{solv},1}) \\ +\beta_{\text{solv}}\left(\frac{i_s}{F} - \frac{dL_{\text{sei}}}{dt}c_{\text{solv},1}\right), & \text{if } i = 1 \\ \alpha_{\text{solv}}(c_{\text{solv},i+1} - 2c_{\text{solv},i} + c_{\text{solv},i-1}) \\ +\gamma_{\text{solv}}(c_{\text{solv},i+1} - c_{\text{solv},i-1}), & \text{if } 1 < i < N_{\text{sei}} \\ 0, & \text{if } i = N_{\text{sei}} \end{cases}$$
(20)

where $\alpha_{solv} = ((D_{solv}(T_c))/(L_{sei}\Delta\xi)^2), \quad \gamma_{solv} = (((\xi - 1)/(2L_{sei}\Delta\xi))(dL_{sei}/dt)), \text{ and } \beta_{solv} = ((2/(L_{sei}\Delta\xi)) + (1/(D_{solv}(T_c)))(dL_{sei}/dt)).$

Here, $\boldsymbol{c}_{\text{solv}} = [c_{\text{solv},1}, \dots, c_{\text{solv},N_{\text{sei}}}]^T \in \mathbb{R}^{N_{\text{sei}}}$ with $c_{\text{solv},1} = c_{\text{solv}}^{\text{surf}}$ and $\boldsymbol{\xi} = ((r - R_{s,n})/L_{\text{sei}})$ and $\Delta \boldsymbol{\xi} = (1/(N_{\text{sei}} - 1))$ with N_{sei} as the number of SEI layer discretization points [25].

E. Surrogate Model for Solvent Diffusion Dynamics

In the cell model, the aging dynamics, inclusive of the SEI layer growth and solvent diffusion, acts as the slow dynamics. In particular, the characteristic time scales of the battery dynamics can be calculated as [17]

$$t_{\text{ter}} = \frac{R_{\text{cell}}^2}{\phi}, \quad t_{\text{elec}} = \frac{R_{s,n}^2}{D_{s,n}}, \quad t_{\text{ag}} = \frac{R_{s,n}^2}{D_{\text{solv}}}$$
(21)

where t_{ter} , t_{elec} , and t_{ag} are the time scales of the thermal, electrochemical, and aging dynamics, respectively, R_{cell} is the radius of a cylindrical lithium-ion cell, ϕ is the thermal diffusivity, $R_{s,n}$ is the particle radius in the negative electrode, $D_{s,n}$ is the solid-phase diffusion in the negative electrode, and D_{solv} is the solvent diffusion. Incorporating parameter values from the literature [17], [31], [32] shows that t_{ag} is in the order of 10^8 s, while t_{ter} and t_{elec} are in the orders of 10–100 and 10^3 s, respectively, implying that the cell model is a threetime-scale system in which $t_{\text{ter}} < t_{\text{elec}} \ll t_{\text{ag}}$.

The difference in temporal scales in the cell dynamics is the cause of long—at time, prohibitive—simulation times that are not compatible with the design of an optimization strategy. In the aging dynamics, the SEI layer growth is the low-dimensional slow variable whose dimension is determined



Fig. 2. Scheme of the surrogate model derivation to calculate c_{solv}^* .

by the number of cells in the battery modules, whereas the solvent concentration dynamics is a high-dimensional state whose dimension is dependent on the number of discretization points of the solvent diffusion PDE.

The integration of solvent diffusion dynamics (10) represents the major bottle neck from a computational standpoint. To get a fast simulation time, we propose a surrogate model to capture the solvent diffusion dynamics (20) based on a joint optimization/curve fitting approach (see Fig. 2). The surrogate model is built to identify a constant value of c_{solv} as a function of I_{cell} and T_{amb} to ensure that the final value of the SEI layer thickness from the high-fidelity model is accurately predicted. Note that the solvent concentration c_{solv}^{surf} is used to calculate the side-reaction current (13) based on which SEI layer growth and cell capacity loss are calculated [see (12)].

The following unconstrained optimization problem is formulated to find the optimal c_{solv}^{surf*}

$$c_{\text{solv}}^{\text{surf}*} = \min_{c_{\text{solv}}} \left\| L_{\text{sei}}^{\text{hf}} - L_{\text{sei}}^{\text{lf}} (c_{\text{solv}}^{\text{surf}}) \right\|$$
(22)

where L_{sei}^{hf} is the SEI layer thickness from the SPM inclusive of the solvent diffusion model (20), whereas $L_{sei}^{lf}(c_{solv}^{surf})$ is the SEI layer thickness when the constant solvent concentration is used. Note that the SEI layer thickness values L_{sei}^{hf} and $L_{\rm sei}^{\rm lf}(c_{\rm solv}^{\rm surf})$ are the final values at the end of the charging time. In Fig. 3, the difference between the final SEI layer thickness values from the SPM with solvent diffusion dynamics $L_{\text{sei}}^{\text{hf}}$ and the SEI layer thickness from the surrogate model $L_{\text{sei}}^{\text{lf}}(c_{\text{soly}}^{\text{surf}*})$ is shown for six different charging C-rates of c_{solv}^{a} (c_{solv}^{surf*}) is shown for six different charging C-rates of [3C, 4C, 5C, 6C, 7C, 8C] at three different ambient temperatures $T_{\text{amb}} = [15 \text{ °C}, 25 \text{ °C}, 35 \text{ °C}]$. As observed, the SEI layer thickness values coincide with each other, thereby proving that the surrogate model is a suitable choice to replace the higher dimensional model to solve the OCP successfully with lower computation cost. The resulting optimal values of c_{solv}^{surf*} obtained from the unconstrained optimization problem are fit as a function of I_{cell} and T_{amb} using the 5th-order polynomials.

F. State-Space Representation: Module-Level

The state and parameter heterogeneity due to manufacturing imperfections and nonuniform operating conditions can cause exacerbated aging of the battery pack when compared to a single cell. The overall thermal and aging effects of the cells in a module can be captured through heat transfer between



Fig. 3. Comparison of the SEI layer thickness values between the SPM with solvent diffusion dynamics and the proposed surrogate model at the end of charging cycles at C-rates = [3C, 4C, 5C, 6C, 7C, 8C] at three different ambient temperatures $T_{amb} = [15 \text{ °C}, 25 \text{ °C}, 35 \text{ °C}].$ TABLE I

cells. The thermal interconnection between adjacent cells in the battery module is provided via the thermal resistance term R_m among a cell k with the downstream and upstream cells, k - 1 and k + 1, respectively. This results in surface temperature dynamics of interconnected cells that are modeled as follows [33]:

$$C_{s}\frac{dT_{s_{k}}}{dt} = \frac{T_{\text{amb}} - T_{s_{k}}}{R_{u}} - \frac{T_{s_{k}} - T_{c_{k}}}{R_{c}} + \frac{T_{s_{k}} - T_{s_{k+1}}}{R_{m}} + \frac{T_{s_{k}} - T_{s_{k-1}}}{R_{m}}.$$
 (23)

The core temperature of cell k is resolved using the relation already stated in (9). In the module-level matrix $A_{\text{therm}}^{\text{mod}}$ in Table I, the surface temperature states, T_{s_k} , embed the cell-to-cell heat transfer from (23).

A convenient shorthand term for module-level dynamics with N_{cell} series-connected cells is given as follows:

$$\dot{\boldsymbol{c}}_{s,j}^{\text{mod}} = A_{s,j}^{\text{mod}} \boldsymbol{c}_{s,j}^{\text{mod}} + B_{s,j}^{\text{mod}} \boldsymbol{u} - \boldsymbol{G}_{s,j}^{\text{mod}}$$

$$\dot{\boldsymbol{T}}^{\text{mod}} = A_{\text{therm}}^{\text{mod}} \boldsymbol{T}^{\text{mod}} + B_{\text{therm}}^{\text{mod}} \boldsymbol{u} + \boldsymbol{G}_{\text{therm}}^{\text{mod}} \boldsymbol{T}_{\text{amb}}$$

$$\dot{\boldsymbol{L}}_{\text{sei}}^{\text{mod}} = \boldsymbol{G}_{\text{sei}}^{\text{mod}}$$

$$\dot{\boldsymbol{\varrho}}^{\text{mod}} = \boldsymbol{G}_{\varrho}^{\text{mod}}$$

$$\dot{\boldsymbol{c}}_{\text{solv}}^{\text{mod}} = \boldsymbol{G}_{\text{solv}}^{\text{mod}}$$
(24)

where u includes the currents of all cells, the module-level block diagonal coefficient matrices and state vectors are listed in Table I, and the module state vector at the system level is

$$\boldsymbol{z}(t) = \left[\boldsymbol{c}_{s,j}^{\text{mod}} \ \boldsymbol{T}^{\text{mod}} \ \boldsymbol{L}_{\text{sei}}^{\text{mod}} \ \boldsymbol{Q}^{\text{mod}} \ \boldsymbol{c}_{\text{solv}}\right]^{T}.$$
 (25)



Fig. 4. Battery module with N_{cell} series-connected cells, where each cell is connected to balancing hardware. For both schemes (OCP-SCT and OCP-DCT), the proposed optimal controller realized through NLP using direct collocation optimizes the variables annotated in blue for fast charging and minimized degradation.

Note that the RHS of the solvent diffusion dynamics, $\mathbf{G}_{\text{solv}}^{\text{mod}}$, is a nonlinear function of the states and input that can be derived for each cell using (20). It should be also pointed out that $\alpha_{s,j}$ and $\beta_{s,j}$ used in $A_{s,j}^{\text{mod}}$, $B_{s,j}^{\text{mod}}$, and $\mathbf{G}_{s,j}^{\text{mod}}$ vary between cells due to cell heterogeneity of design parameters, nonuniform aging, and temperature distribution, respectively.

Upon the creation of the surrogate model to replace the solvent diffusion dynamics, presented in Section III-E, the module state vector used in the proposed optimal control design is given by³

$$\boldsymbol{x}(t) = \begin{bmatrix} \boldsymbol{c}_{s,j}^{\text{mod}} \ \boldsymbol{T}^{\text{mod}} \ \boldsymbol{L}_{\text{sei}}^{\text{mod}} \ \boldsymbol{Q}^{\text{mod}} \end{bmatrix}^T \in \Re^{N_s}$$
(26)

where the number of states is $N_s = N_{cell}(4 + 2(N_r - 1))$ for a given N_r .

IV. OCP FORMULATION

In this section, we formulate a multiobjective optimal control framework for fast charging and minimum degradation of a battery module with N_{cell} series-connected imbalanced cells, as shown in Fig. 4. In this configuration, the module capacity is limited by the capacity of the weakest cell in the string. In this work, we account for the intrinsic heterogeneity among the cells in terms of charge, temperature, and SOH. Moreover, battery health is defined both in terms of Q and R_{sei} , both dependent on L_{sei} as seen from (7) and (19). To model cells subject to SOH imbalances, the selection of different initial conditions for L_{sei} is made.

A multiobjective OCP is formulated for two different charging schemes, OCP-SCT and OCP-DCT. The former assumes that all cells are charged simultaneously, irrespective of their nonhomogeneous initial states, whereas the latter assumes different times of charging of the cell to reflect the nonuniform initial states the cells are at. In particular, OCP-DCT is aimed at providing a charging strategy that extends the battery life and provides more flexibility against heterogeneity among the cells. From Fig. 4, $I_0 = I_{cell_k} + I_{B_k}$ for $k = 1, ..., N_{cell}$ from which one can define the vector

$$I_{\text{cell}} = [I_{\text{cell}_1}, \dots, I_{\text{cell}_{N_{\text{cell}}}}]^T = [I_0 - I_{B_1}, \dots, I_0 - I_{B_{N_{\text{cell}}}}]^T.$$
(27)

³Note that c_{solv} included in z(t) is now excluded from the system-level state vector x(t) due to the inclusion of the surrogate model.

During charging, the module current $I_0 \in \Re$ and the vector of balancing currents $I_B = [I_{B_1}, \ldots, I_{B_{N_{cell}}}]^T \in \Re^{N_{cell}}$ are unknown and optimally planned. Each cell k is connected in parallel to an active balancing circuitry whose current I_{B_k} is determined by the proposed optimal controller. The following OCP for the OCP-DCT scheme is formulated as:

$$X^* = \underset{X \in \mathfrak{N}^{N_{\text{opt}}}}{\operatorname{argmin}} \alpha \beta_1 h(t_f) + (1 - \alpha) \left(\beta_2 g_1(L_{sei}) + \beta_3 g_2(\dot{L}_{sei}) \right)$$
(28)

where the vector of optimization variables X is comprised of the vector of final times of charging $t_f = [t_{f_1}, \ldots, t_{f_{N_{\text{cell}}}}]^T \in \Re^{N_{\text{cell}}}$, and the system state $x(t) \in \Re^{N_s}$, the module current I_0 , and the balancing current vector I_B

$$\boldsymbol{X} = \begin{bmatrix} \boldsymbol{t}_f, \boldsymbol{x}(t), I_0(t), \boldsymbol{I}_B(t) \end{bmatrix}^T \in \mathfrak{R}^{N_{\text{opt}}}.$$
 (29)

The number of optimization variables is $N_{\text{opt}} = N_s + 2N_{\text{cell}} + 1$ and the continuously differentiable functions g_1 , g_2 , and h are defined as

$$g_1(L_{sei}) = \frac{1}{N_{cell}} \sum_{k=1}^{N_{cell}} L_{sei_k}$$

$$g_2(\dot{L}_{sei}) = \frac{1}{N_{cell}} \sum_{k=1}^{N_{cell}} \dot{L}_{sei_k}$$

$$h(t_f) = \frac{1}{N_{cell}} \sum_{k=1}^{N_{cell}} t_{f_k}.$$
(30)

Note that $g_1(L_{sei})$, $g_2(\dot{L}_{sei})$, and $h(t_f)$ are operators that return the average of SEI layer thicknesses at the end of charging, the average of the SEI layer thickness growth rates, and the average of charging times, respectively. Thus, the OCP (28) along with the definitions (30) forms a min-mean optimization problem. The positive scalars β_1 [s⁻¹], β_2 [sm⁻¹], and β_3 [sm⁻¹] are optimization weights corresponding to the charging time and SEI layer growth objectives, which are chosen prior to our exploration of the parameter space to set the objective terms on the same order of magnitude. The dimensionless scalar $0 \le \alpha \le 1$ is a tradeoff coefficient that can be adjusted to give three different paradigms: fast charging ($\alpha = 1$), minimum degradation ($\alpha = 0$), and balanced charging degradation ($0 < \alpha < 1$), as demonstrated in Section VI-D.

The operation of the battery module is subject to the dynamic constraints (24) and the following operating constraints for each cell with $k = 1, ..., N_{cell}$. To establish safety metrics, module and balancing currents, voltages, core and surface temperatures, and solid concentrations of all cells are enforced to lie within their physical bounds for $k = 1, ..., N_{cell}$

$$I_{B_{\min}} \leq I_{B_k}(t) \leq I_{B_{\max}}, \quad I_{0_{\min}} \leq I_0(t) \leq I_{0_{\max}}$$

$$V_{\text{cellmin}} \leq V_{\text{cell}_k}(t) \leq V_{\text{cellmax}}$$

$$T_{lk_{\min}} \leq T_{lk}(t) \leq T_{lk_{\max}}, \quad l \in \{c, s\}$$

$$\theta_{0\%}^{j} c_{s,jk_{\max}} \leq c_{s_jk}(t) \leq \theta_{100\%}^{j} c_{s,jk_{\max}}, \quad j \in \{n, p\}. \quad (31)$$

Initial conditions of the states are taken into consideration as equality constraints

$$L_{\text{sei}_k}(t_0) = L_{\text{sei}_{0_k}}, \quad Q_k(t_0) = Q_{0_k}$$

$$T_{lk}(t_0) = T_{lk_0}, \quad l \in \{c, s\}$$
(32)

$$SOC_k(t_0) = SOC_{initial_k}$$
 (33)

and cells are charged to the same targeted SOC

$$SOC_k(t_{f_k}) = SOC_{target}.$$
 (34)

In the OCP-DCT scheme, charging time is allowed to be different for each cell and an upper bound on the charging time is imposed as well

$$0 \le t_{f_k} \le t_{f_{\max}}.\tag{35}$$

A second problem in the context of optimal charging is investigated in this work where simultaneous charging time of all cells in the module must be achieved. We refer to this formulation as the OCP-SCT.

The OCP-SCT problem resembles the OCP-DCT scheme except for the following differences.

- The final times of charging are the same for all cells from which one can consider t_f ∈ ℜ.
- 2) The number of optimization variables reduces to $N_{\text{opt}} = N_s + N_{\text{cell}} + 2$; hence,

$$\boldsymbol{X} = \left[t_f, \boldsymbol{x}(t), I_0(t), \boldsymbol{I}_{\boldsymbol{B}}(t)\right]^T \in \Re^{N_{\text{opt}}}$$
(36)

in which t_f is now a scalar.

- 3) The cost function associated with charging in (28) reduces to $h(t_f) = t_f$.
- 4) The constraint associated with the charging time reduces to $0 \le t_f \le t_{f_{\text{max}}}$ in which the charging time of all cells is the same.

In Section V, we solve the OCP-SCT and OCP-DCT subject to dynamic constraints (24) and the operating constraints (31)–(35).

Remark 1: Note that the vector of optimization variables X proposed in (29) and (36) considers I_0 to be a free variable. Only a portion of the module current I_0 provided by the optimal solution flows through the cells (I_{cell}), meaning that some of I_0 is wasted (by bleeding through the balancing hardware). However, since the value of I_0 does not directly affect the objective functions in (28), the resulting optimal solution in terms of fast charging while ensuring minimum degradation is guaranteed. The rationale behind having I_0 and I_B in the optimization variable X is to build on it in the future work by including additional objective functions such as follows:

- 1) minimization of module energy consumption (which is dependent on I_0).
- 2) minimization of the temperature or heat loss in any general balancing hardware (which is dependent on I_B).

In the proposed formulation, since the objective functions are not explicitly minimizing the energy consumption, the value of I_0 is not the optimal controller's immediate concern. To that end, there exists an alternate formulation of the optimization variable vector X that can potentially be used to solve the same OCP by eliminating I_0 and I_B from the vector of optimization variables and replace it with the cell currents I_{cell} , given by

$$\boldsymbol{X} = \begin{bmatrix} \boldsymbol{t}_{f}, \boldsymbol{x}(t), \boldsymbol{I}_{cell}(t) \end{bmatrix}^{T}$$
(37)

2) that satisfy the following algebraic relationships:

$$I_0 = \min(I_{\text{cell}_1}, \dots, I_{\text{cell}_{N_{\text{cell}}}})$$
$$I_B = [I_0 - I_{\text{cell}_1}, \dots, I_0 - I_{\text{cel}_{N_{\text{cell}}}}].$$
(38)

This formulation results in one less optimization variable (without the I_0 variable), and however, it may not be preferred in the future when objective functions penalizing module energy consumption and temperature or heat loss in the balancing hardware are to be incorporated.

V. OPTIMAL CONTROL ALGORITHM

In this article, the direct collocation method [26] is employed to solve the OCP characterized by nonlinear coupled dynamic constraints (24). The original OCP (28) is transcribed into an NLP problem [27] by approximating all elements of the unknown vector X with polynomial splines. Spline approximation refers to the operation of replacing a continuous trajectory with a sequence of polynomial segments that are glued together at given BPs.

This results in all trajectories to be discretized in time $0 = t_0 < t_1 < \cdots < t_{N_{BP}} = t_f$, where N_{BP} is the number of BPs and t_0 and t_f are the initial and final times, respectively. The order of polynomial segments, d, and the degree of smoothness over the BPs, s, are specified in such a way that the continuity of discretized trajectories at BPs and between them is ensured. A spline can be parameterized as the weighted sum of B-splines—piecewise polynomials of order d—such that each optimization variable vector can be approximated as

$$X_p(t) = \sum_{q=1}^{N_{\text{FP}_p}} \mathcal{B}_{p,q} \omega_{p,q}, \text{ for } p = 1, \dots, N_{\text{opt}}$$
(39)

where $\mathcal{B}_{p,q}$ and $\omega_{p,q}$ are the *q*th B-spline and free parameters of the *p*th optimization variable, respectively, and $N_{\text{FP}_p} = N_P(d_p - s_p) + s_p$ is the number of free parameters for the *p*th optimization variable with $N_P = N_{\text{BP}} - 1$ as the number of polynomial segments [34]. By parameterizing all of the system trajectories t_f , x(t), $I_0(t)$, and $I_B(t)$ (t_f is scalar in case of OCP-SCT), the total number of free parameters is calculated as

$$N_{\rm FP}^{t} = N_{\rm FP_{x}} N_{s} + (N_{\rm FP_{I_{R}}} + N_{\rm FP_{t_{f}}}) N_{\rm cell} + N_{\rm FP_{I_{0}}}$$
(40)

where N_{FP_x} , $N_{\text{FP}_{I_B}}$, $N_{\text{FP}_{I_0}}$, and $N_{\text{FP}_{I_f}}$ are the numbers of free parameters for each state, balancing and module currents, and charging times, respectively. These are design parameters to be selected by users.

With this approximation in hand, the original OCP (28) is transcribed to the NLP problem as follows:

$$\mathbf{P}^* = \underset{P}{\operatorname{argmin}} J(P)$$

s.t. $g_{P_1}(P) = \mathbf{0}, \ g_{P_2}(P) \leq \mathbf{0}, \ P_{\min} \leq P \leq P_{\max}$ (41)

where $\boldsymbol{P} = [\omega_{p,q}] \in \mathfrak{R}^{N_{\text{fp}}^{t}}$ is the finite set of free parameters; and $J \in \mathfrak{R}$, and $\boldsymbol{g}_{P_1} \in \mathfrak{R}^{m_1}$ and $\boldsymbol{g}_{P_2} \in \mathfrak{R}^{m_2}$ are the cost, and the vectors of linear/nonlinear equality and inequality constraints, respectively, all expressed in terms of the vector of the static parameters \boldsymbol{P} . The Lagrangian function $\mathcal{L} : \mathfrak{R}^{N_{\text{FP}}^{t}} \times \mathfrak{R}^{m_{1}} \times \mathfrak{R}^{m_{2}} \to \mathfrak{R}$ associated with the NLP problem (41) is defined as

$$\mathcal{L}(\boldsymbol{P},\boldsymbol{\mu}_1,\boldsymbol{\mu}_2) = J(\boldsymbol{P}) + \boldsymbol{\mu}_1^T \boldsymbol{g}_{\boldsymbol{P}_1}(\boldsymbol{P}) + \boldsymbol{\mu}_2^T \boldsymbol{g}_{\boldsymbol{P}_2}(\boldsymbol{P}) \quad (42)$$

with $\mu_1 \in \Re^{m_1}$ and $\mu_2 \in \Re^{m_2}$. The KKT optimality conditions [35] associated with (42) are

$$\nabla \mathcal{L} = \nabla J(\boldsymbol{P}^{*}) + \sum_{r=1}^{m_{1}} \mu_{1_{r}}^{*} \nabla g_{P_{1_{r}}}(\boldsymbol{P}^{*}) + \sum_{r=1}^{m_{2}} \mu_{2_{r}}^{*} \nabla g_{P_{2_{r}}}(\boldsymbol{P}^{*}) = 0 \quad \text{(Stationarity)}$$
(43)

 $g_{P_1}(P^*) = 0$, for $r = 1, ..., m_1$

$$g_{P_2}(P^*) \le 0$$
, for $r = 1, \dots, m_2$ (Primal feasibility) (44)

$$\mu_{2_r}^* \ge 0$$
, for $r = 1, \dots, m_2$ (Dual feasibility) (45)

$$\sum_{r=1}^{m_2} \mu_{2_r}^* g_{P_{2_r}}(\boldsymbol{P}^*) = 0 \quad \text{(Complementary slackness)} \tag{46}$$

where conditions (43)–(46) are called stationarity, primal feasibility, dual feasibility, and complementary slackness, respectively, and μ_{1_r} for $r = 1, \ldots, m_1$ and μ_{2_r} for $r = 1, \ldots, m_2$ are KKT multipliers. For any continuously differentiable cost J and constraints g_{P_1} and g_{P_2} , if there exists a pair of $(\boldsymbol{\mu}_1^*, \boldsymbol{\mu}_2^*)$ such that the KKT conditions (43)–(46) hold, then a solution \boldsymbol{P}^* is a local optimum for the NLP problem (41). It should be pointed out that when \boldsymbol{P}^* and $(\boldsymbol{\mu}_1^*, \boldsymbol{\mu}_2^*)$ are any primal–dual optimal points with zero duality gap (strong duality), and then, any pair of $(\boldsymbol{P}^*, (\boldsymbol{\mu}_1^*, \boldsymbol{\mu}_2^*))$ satisfies the KKT conditions (43)–(46) [35].

Under the direct collocation approach, the cost and constraints are applied to the optimization variables t_f , x(t), $I_0(t)$, and $I_B(t)$ (t_f is scalar in case of OCP-SCT) at CPs. In this article, we determine the CPs based on the GQF using which the BPs do not coincide with the CPs necessarily. GQF can find an optimal set of CPs (not equally spaced) to fit high-degree polynomials. After transcription of the OCP to the NLP problem using the direct collocation, the interior point solver IPOPT [28] is employed to solve the NLP problem. All the dynamics, operating constraints, and the cost are implemented symbolically. This formulation provides symbolic differentiation of the OCP, which in turn results in remarkable improvement in convergence time and solving feasibility.

Remark 2: In view of (40), the number of free parameters reduces to $N_{\text{FP}}^t = N_{\text{FP}_x}N_s + N_{\text{FP}_{I_B}}N_{\text{cell}} + N_{\text{FP}_{I_f}} + N_{\text{FP}_{I_0}}$ when the OCP-SCT scheme is selected for the OCP. This results in the NLP with less parameters to be optimized with a reduction of computational effort and convergence time.

VI. SIMULATION RESULTS

In this section, we test the effectiveness of the proposed optimal control algorithm for both OCP-SCT and OCP-DCT schemes on a battery module with two series-connected imbalanced cells (i.e., $N_{cell} = 2$), where each cell is connected in parallel to an active balancing circuitry (see Fig. 4).

TABLE II Specifications of the Cylindrical 18650 LIB Cell Used in the Simulations

Model	Specification (Sony VTC4)
Cathode chemistry	NMC
Anode chemistry	Graphite
Nominal capacity	2 Ah
Nominal voltage	3.6 V
Minimum voltage	2.5 V
Maximum voltage	4.2 V
2	1



Fig. 5. Open-circuit potentials of NMC cathode/graphite anode cell.

A. Initialization and Setup

The battery considered in this article is a cylindrical 18650, 2-Ah lithium-ion NMC cathode/graphite anode cell whose characteristics are reported in Table II [17]. The open-circuit potentials of each electrode, U_j , in terms of the surface stoichiometry, $\theta_j^{\text{surf}} = c_{s,j}^{\text{surf}}/c_{s,j}^{\text{max}}$, are shown in Fig. 5. Throughout the simulations, we assume that there is an initial SOC imbalance among the cells [SOC₁(0) \neq SOC₂(0)], while no mismatch between temperature, SEI layer thickness, resistance, and capacities of individual battery cells is assumed.

The physical bounds for the operating constraints (31)–(35)are set to $I_{B_{\min}} = -6$ A, $I_{0_{\min}} = -16$ A, $I_{B_{\max}} = 0$ A, $I_{0_{\max}} =$ -12 A, SOC_{target} = 0.8, $V_{cellmin} = 2.5$ V, $V_{cellmax} = 4.2$ V, $t_{f_{\text{max}}} = 2000 \text{ s}, \ \tilde{T}_{lk_{\text{min}}} = 5 \text{ °C}, \text{ and } T_{lk_{\text{max}}} = 45 \text{ °C} \text{ with } l \in \{c, s\}$ and k = 1, 2. Note that the minimum and maximum voltages follow the battery specifications mentioned in Table II. The initial conditions are picked as $L_{sei_k}(0) = 5 \times 10^{-9}$ m (this is the typical SEI later thickness observed for a fresh cell), $Q_k(0) = 2$ Ah, and $T_{ck}(0) = T_{sk}(0) = T_{amb}$. The numbers of discretization points are set to $N_{r,i} = N_{sei} = 10$. Thus, the number of states used in the OCP is $N_s = 44$. Given the nominal capacity $Q_{\text{nom}} = Q_k(0) = 2$ Ah, the selected bounds for module and balancing currents result in having cell current between -16 and -6 A. For the balanced chargingdegradation scenario, the optimization weights and the tradeoff coefficients are selected to be $\beta_1 = 1$ [s⁻¹], $\beta_2 = \beta_3 = 5 \times$ 10^8 [sm⁻¹], and $\alpha = 0.5$; they are the same for both the OCP-SCT and the OCP-DCT schemes.

For the surrogate model development, the cell current is discretely sampled within its range, i.e., [-16 - 6] A (with a sampling current of 2 A), and ambient temperatures are chosen to be [15, 25, 35] °C. The 5th-order polynomials are fit to the optimal points $c_{\text{solv}}^{\text{surf*}}$ for all six sampled currents and each ambient temperature. The MATLAB built-in functions fminsearch and polyfit are employed to solve the optimization (22) and fit the polynomials, respectively.



Fig. 6. Results from the multiobjective OCP-SCT scheme with initial SOC mismatch SOC(0) = [0.2, 0.4] at $T_{amb} = 25$ °C.



Fig. 7. Results from the multiobjective OCP-DCT scheme with initial SOC mismatch SOC(0) = [0.2, 0.4] at $T_{amb} = 25$ °C.

B. Initial SOC Mismatch With Different Ambient Temperatures

At an ambient temperature of $T_{\rm amb} = 25$ °C, we first solve OCP-SCT and OCP-DCT when the initial SOC for the two cells is set to SOC(0) = [0.2, 0.4]. From Fig. 6, under OCP-SCT scheme, both cells are charged simultaneously, while their voltages lie within $V_{\rm cellmin} = 2.5$ V and $V_{\rm cellmax} = 4.2$ V. In Cell 1 (with lower initial SOC), there is a higher rate of charge than in Cell 2 and the same time of charge is enforced. This, in turn, leads to Cell 1 to experience more aging and achieve higher core and surface temperature, as it absorbs more current (the lower I_{B_1} results in the higher I_{cell_1}).

On the contrary, charging times are different for Cells 1 and 2 when using the OCP-DCT scheme, as shown in Fig. 7. As expected, Cell 2 (at higher initial SOC) is charged faster, while both cells have the same rates of charging across the different ambient temperatures. Once Cell 2 is fully charged at t_{f_2} , SOC₂ is kept constant until Cell 1 reaches SOC_{target} at t_{f_1} ; the cell current is absorbed by the power units implementing the active balancing circuitry, leading to $I_0 = I_{B_2}$ over $t_{f_2} \le t \le t_{f_1}$; V_{cell_2} drops at t_{f_2} and



Fig. 8. Optimization results for (a) OCP-SCT and (b) OCP-DCT schemes based on 20 simulations with initial SOC mismatch taken from a uniform distribution over the interval [0.2, 0.4] at different ambient temperatures $T_{amb} = [15, 25, 35]$ °C.



Fig. 9. Optimization results for (a) OCP-SCT and (b) OCP-DCT schemes based on 20 simulations with initial L_{sei} mismatch taken from a uniform distribution over the interval [4, 6] × 10⁻⁹ m at different ambient temperatures $T_{amb} = [15, 25, 35]$ °C.

TABLE III PERFORMANCE COMPARISON BETWEEN THE OCP-SCT AND OCP-DCT SCHEMES WITH INITIAL SOC MISMATCH SOC₁(0) = 0.2 and SOC₂(0) = 0.4 at DIFFERENT AMBIENT TEMPERATURES T_{amb} = [15, 25, 35] °C. THE BEST VALUE OF EACH METRIC IS SHOWN IN BOLD

T_{amb}	Scheme	$(\Delta L_{sei_1}, \Delta L_{sei_2})\%$	$(\Delta Q_1, \Delta Q_2)\%$	(t_{f_1}, t_{f_2}) s
$15^{\circ}C$	OCP-SCT	(0.079, 0.021)	$(20, 5.5) \times 10^{-5}$	(406, 406)
	OCP-DCT	(0.021, 0.021)	$(5.43, 5.41) \times 10^{-5}$	(571, 386)
$25^{\circ}C$	OCP-SCT	(9.97, 6.13)	(0.025, 0.015)	(463, 463)
	OCP-DCT	(5.95, 5.59)	(0.015, 0.014)	(595, 393)
$35^{\circ}C$	OCP-SCT	(60, 47)	(0.15, 0.12)	(471 , 471)
	OCP-DCT	(38, 29)	(0.098, 0.073)	(671, 458)

remains constant over $t_{f_2} \le t \le t_{f_1}$; the core and surface temperatures of Cell 2 start decreasing at t_{f_2} ; and the rates of L_{sei_2} and Q_2 slow down after t_{f_2} . The figures also show that the OCP-DCT scheme reduces degradation gradient between cells [i.e., $L_{sei_2}(t_{f_2}) - L_{sei_1}(t_{f_1})$] at all different ambient temperatures.

To compare the results of OCP-SCT and OCP-DCT, Table III lists quantitative comparisons between the two schemes at $T_{amb} = [15, 25, 35]$ °C. Referring to this table, under either OCP-SCT or OCP-DCT, when T_{amb} increases, the following trends are inferred: 1) the SEI layer thickness variation of both cells increases; 2) the capacity loss variation of each cell increases; and 3) the charging time of each cell increases. According to this table, OCP-DCT decreases max($\Delta L_{sei_1}, \Delta L_{sei_2}$) by 73%, 40%, and 40% over OCP-SCT when T_{amb} is set to 15 °C, 25 °C, and 35 °C, respectively; this, in turn, leads to max($\Delta Q_1, \Delta Q_2$) to be decreased by 72%, 40%, and 35% when OCP-DCT is used. In terms of charging time, however, OCP-DCT increases $\max(t_{f_1}, t_{f_2})$ by 40%, 28%, and 42% over OCP-SCT when T_{amb} is 15 °C, 25 °C, and 35 °C, respectively.

C. Robustness to Initial SOC and SOH Imbalances

To further elaborate on the robustness of the proposed OCP-SCT and OCP-DCT schemes, this section is devoted to perform multiple simulations at different ambient temperatures [15, 25, 35] °C for initial values of SOC and SOH imbalance randomly taken from uniform distributions.

1) Random Initial SOC Imbalance: For each ambient temperature, $N_{\rm sim} = 20$ simulations are carried out where initial SOCs are drawn from a uniform distribution over the interval [0.2, 0.4]. It can be seen from Fig. 8 that under either scheme, when the ambient temperature increases, the maximum of SEI layer thickness variations of the cells max($\Delta L_{\rm seil}^*$, $\Delta L_{\rm sei}^*$) and



Fig. 10. Pareto fronts for (a) OCP-SCT and (b) OCP-DCT schemes at different ambient temperatures $T_{amb} = [15, 25, 35]$ °C when the optimization tradeoff coefficient is discretely sampled as $\alpha = \{0, 0.25, 0.5, 0.75, 1\}$ and with an initial SOC mismatch SOC(0) = [0.2, 0.4].

the maximum charging times of the cells $\max(t_{f_1}^*, t_{f_2}^*)$ increase as well.

Numerical results show that under the OCP-SCT scheme, $\max_{k_{N_{\text{sim}}}}(\Delta L^*_{\text{sei}_1}, \Delta L^*_{\text{sei}_2})$ max = [0.08, 9.62, 57.15]% and $\max_{k_{N_{sim}}}(t_f^*) = [510, 596, 717]$ s for $T_{amb} = [15, 25, 35]$ °C, where $k_{N_{sim}} = 1, ..., 20$ is the $k_{N_{sim}}$ th simulation. On the other hand, with an OCP-DCT and under different ambient temperatures, scheme max $\max_{k_{N_{\text{sin}}}} (\Delta L^*_{\text{sei}_1}, \Delta L^*_{\text{sei}_2}) = [0.04, 6.22, 38.64]\%$ and max $\max_{k_{N_{sim}}}(t_{f_1}^*, t_{f_2}^*) = [587, 646, 738]$ s. These findings are in agreement with our observations in Table III, showing that the optimization under the OCP-DCT scheme leads to the battery module with lower variation of SEI layer thickness and longer charging time regardless of the ambient temperature at which the simulation is performed.

2) Random Initial SOH Imbalance: In this experiment, $N_{\rm sim} = 20$ simulations are run for each ambient temperature for both control schemes, where in each simulation, initial L_{sei} values are drawn from a uniform distribution over the interval $[4, 6] \times 10^{-9}$ m to represent the SOH imbalance at the beginning of the battery life. From Fig. 9, the results reveal that with the OCP-SCT scheme, $\max \max_{k_{N_{\rm sim}}} (\Delta L^*_{\rm sei_1}, \Delta L^*_{\rm sei_2}) = [0.09, 14.94, 77.10]\%$ and $\max_{k_{MC}}(t_f^*) = [498, 547, 783]$ s, and under the OCP-DCT scheme, max $\max_{k_{N_{sim}}} (\Delta L_{sei_1}^*, \Delta L_{sei_2}^*) = [0.06, 9.98, 57.87]\%$, and max $\max_{k_{N_{sim}}} (t_{f_1}^*, t_{f_2}^*) = [546, 620, 815]$ s all for $T_{amb} =$ [15, 25, 35] °C. These results are in line with what we found from the case of initial SOC imbalance, showing that the OCP-DCT scheme is able to mitigate the variation of SEI layer thickness at the cost of higher charging time irrespective of the ambient temperature. In comparison with the case of initial SOC imbalance, the simulations with initial SOH imbalance lead to the battery module with higher variation of SEI layer thickness at any ambient temperature used.

D. Pareto Fronts: Effect of Tradeoff Coefficient a

Recall that the optimization tradeoff coefficient was picked to be $\alpha = 0.5$ in Section VI-A to study the balanced chargingdegradation scenario. However, this parameter could be varied to weigh more or less battery degradation over time of charge, given that the two costs have conflicting objectives. In this section, α is discretely sampled as $\alpha = \{0, 0.25, 0.5, 0.75, 1\}$ under which OCP-SCT and OCP-DCT schemes are run for different ambient temperatures when there is an initial SOC mismatch SOC(0) = [0.2, 0.4]. Fig. 10 shows that the maximum of SEI layer thickness variations of the cells reduces as α decreases from 1 to 0 at any ambient temperature; the battery module ages less but takes more time for charging when we go from fast charging to minimum degradation objective. This is also supported by numerical results from which when α goes from 1 to 0, at $T_{amb} = [15, 25, 35]$ °C: 1) under OCP-SCT, max $\max_{k_{N_{sim}}} (\Delta L_{sei_1}^*, \Delta L_{sei_2}^*)$ decreases by 72%, 38%, and 23%, and $\max_{k_{N_{sim}}}(t_f^*)$ increases by 66%, 49%, and 75%, respectively, and 2) under OCP-DCT, max $\max_{k_{N_{sim}}} (\Delta L^*_{sei_1}, \Delta L^*_{sei_2})$ decreases by 71%, 26%, and 27%, and max $\max_{k_{N_{sim}}}(t_{f_1}^*, t_{f_2}^*)$ increases by 97%, 46%, and 53%, respectively. Once again, the Pareto fronts support our previously claimed observations, showing that the OCP-DCT scheme can reduce the battery degradation at any ambient temperature tested.

E. Comparison With Conventional Constant Current Profiles

To highlight the advantages and benefits of the proposed optimal controller, a comparison is made with the standard CC charging profiles.⁴ Given that research efforts are underway to enable extreme fast charging, in which the battery pack must be charged to 80% of its capacity in 10–15 min [36], it is reasonable to evaluate the performance of the proposed schemes against higher C-rates (>3C). The candidate CC charging profiles selected are 3C and 8C, which are the minimum and maximum permissible current magnitudes for the cell considered in this work. The two CC profiles along with the OCP-DCT and OCP-SCT profiles proposed in this work are applied to the battery module of two cells connected in series for 300 cycles each.⁵ An initial SOC imbalance of SOC(0) = [0.2, 0.4] is assumed for the two cells in series. The performance of the series-connected cells under the four charging profiles [3C, OCP-DCT, OCP-SCT, 8C] is evaluated at an ambient temperature of 25 °C in terms of charging time for the first cycle and capacity loss at the end of 300 cycles. In this case, the capacity loss for a series-connected cell kis defined as the percentage change in its capacity at the end of 300 cycles, with respect to the nominal capacity, given by $\Delta Q_{\text{loss}}^k = ((Q_{\text{nom}}^k - Q_{300 \text{ cycles}}^k)/Q_{\text{nom}}^k) \times 100\%.$

⁴The CC-CV charging protocol is used in laboratory testing, whereas only CC—or its variants—is used for in-vehicle charging.

 $^{^{5}}$ Note that one cycle is composed of the cells being charged from their initial SOC to the final SOC of 0.8.



Fig. 11. Charging time versus capacity loss tradeoff for (a) Cell 1 and (b) Cell 2, with an initial mismatch of SOC(0) = [0.2, 0.4], when subjected to 3C, OCP-SCT, and 8C profiles for 300 cycles, respectively.

This study intends to demonstrate the health savings each charging strategy offers, in terms of retained capacity over multiple charging cycles.

In Fig. 11(a), we plot the charging time (blue circle) and capacity loss at the end of 300 cycles (red triangle) of Cell 1 for the charging profiles 3C, OPT-DCT, OPT-SCT, and 8C. It is noticed that the charging time reduces as the C-rate increases from 3C to 8C, and as expected, the amount of degradation has the opposite trend in which, as the C-rate increases, the observed capacity loss is higher. However, interestingly, the capacity loss observed for the OCP-SCT and OCP-DCT profiles is lower than 8C and slightly lower than 3C, thereby providing a balanced charging-degradation solution. This indicates that the proposed optimal control profile results in not only minimum degradation compared to both 3C and 8C profiles but also provides a good tradeoff in charging time between the two extremes of 3C and 8C. Similar trends are also observed in the capacity loss of Cell 2 in Fig. 11(b) for all charging profiles. Cell 2 has a higher initial SOC, and hence, its charging time for the OCP-DCT profile is shorter because the scheme allows for DCTs of the cells to account for heterogeneous initial conditions, whereas the charging time of Cell 2 is the same as that of Cell 1 for the OPT-SCT profile. The results validate that the OCP-DCT and OCP-SCT profiles outperform the standard CC profiles by providing a balanced tradeoff between fast charging and minimum degradation. Note that these results are simulated for 300 charging cycles, and however, each cycle only consists of an SOC window from 0.2 or 0.4 to 0.8 (depending on the initial SOC of cells in the module). It follows that as the battery ages and undergoes long-term cycling, the trends and savings, in terms of capacity, will be more pronounced, thereby highlighting the advantages of the proposed optimal controller.

VII. CONCLUSION AND DISCUSSION

A. Conclusion

This article formulated a multiobjective fast charging-minimum degradation OCP for battery modules with N_{cell} series-connected cells with an active balancing circuitry. A surrogate model was proposed to mitigate computational

burden associated with the multi-time-scale nature of the cell dynamics as well as the large-scale nature of LIB modules. Two different OCPs were suggested: OCP-SCT and OCP-DCT. Simulation studies were carried out on a battery module with two series-connected cells in the presence of initial SOC and SOH imbalances under different ambient temperatures. Results demonstrated that both schemes outperform standard CC charging profiles, and degradation and charging time increase as ambient temperature increases. Our findings showed that OCP-DCT provides more flexibility to handle heterogeneities among the cells in terms of obtaining a more uniform degradation among the cells, hence leading to a longer utilization of the module.

In the future, the optimal control of series-connected modules during discharging will be investigated. In the discharging case, the module current I_0 is fixed as per the current/power demand requested by the user or the application, resulting in one less degree of freedom and optimization variable. However, the objective functions will need to be modified according to the discharging scenarios (for instance, charging time objective function is not valid). Having said that, the framework proposed in this article, which consists of using the direct collocation approach to transcribe the OCP into an NLP problem by parameterization of the system states and input, will remain the same. It is worth mentioning that as the number of cells in series increases, the computational burden of solving the OCP will be higher. To that end, the proposed optimal controller is more suited for offline simulations of series-connected cells to generate solutions, trajectories, or reference surface maps, to aid our understanding of the optimal split under different conditions and identify critical conditions or faults. The results from the offline simulations can be used in the form of lookup tables or maps for reference tracking during real-time applications (with reduced-order models) in resource-constrained onboard hardware.

B. Discussion: Impact of Our Work

The adoption of effective active balancing hardware in a battery pack holds the potential to address the issue of guaranteeing longer (>8 years) life when used in EV applications.

Currently, cell balancing via shunt resistors is widely used in the industry. Since the proposed optimal controller is applicable to any general active balancing hardware (either shunt resistors, transistors, or dc/dc converters), it is easier to adopt and it can be immediately deployed without adding additional hardware costs.

In a series-connected module, the capacity of the module is defined by the weakest (most aged) cell. Heterogeneity among cells, if not embraced, will result in some cells to be overly used over time, thus creating a fragile (age-wise) link in the module. The ability to control each single cell while acknowledging their initial states, health, and manufacturing characteristics will result in a module/pack with uniform characteristics and performance. In the quest for solutions that provide longer battery life capability, among discovering new materials and proposing novel manufacturing processes, the system-level solution explored in this article positions itself as an easily deployable method for targeted applications.

ACKNOWLEDGMENT

The authors would like to thank Dr. Won Tae Joe and Dr. Yohwan Choi for providing invaluable guidance for this work.

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