Extending life of Lithium-ion battery packs by taming heterogeneities via an optimal control-based active balancing strategy

Vahid Azimi, Anirudh Allam, and Simona Onori

Abstract—This paper develops a multi-objective fast charging-minimum degradation optimal control problem (OCP) for lithium-ion battery modules, made of series-connected cells, subject to heterogeneity induced by manufacturing defects and non uniform operating conditions, realized via an active balancing circuitry. 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: (i) same-charging-time (OCP-SCT) and (ii) 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. Simulations on an illustrative case study—a battery module with two series-connected cells—are carried out in the presence of intrinsic heterogeneity among the cells 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, 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 the new control.

I. INTRODUCTION

Lithium-ion batteries (LIBs) are the enabling technology to ensure a sustainable future thanks 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]–[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 state of charge 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. Most 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], nonlinear programming (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 battery modules (pack) is that heterogeneity in the parameters among cells (module) is inevitable due to manufacturing and operating conditions which, if not corrected on time, could hinder the performance and longevity of the module (pack) 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, being the cause of differences in temperature, state of charge (SOC), depth of discharge (DOD) and charging rate [14]–[16]. For example, charge imbalance limits the charge/discharge capabilities of the pack, posing limitations on pack-level charge and discharge capabilities; and temperature imbalance 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. 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], wherein each cell in the module is individually managed by the modular converter without the need for equalization circuits.

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 on-board vehicle operation to tackle charge and temperature imbalances by leveraging constrained linear quadratic model predictive control. 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 model predictive control 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 paper, the system under investigation is a Lithium-ion battery (LIB) module of \( N_{\text{cell}} \) Series-connected cells (see Fig. 1), where each cell is connected to an active balancing hardware, which could be either as simple as an active shunt resistor or shunt transistor method or a more sophisticated hardware 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 an active balancing hardware. In Fig. 1, the current of the \( k^{\text{th}} \) cell is given by \( I_{\text{cell}} = I_{0} - I_{B_k} \), where \( I_{0} \) is the module current and \( I_{B_k} \) is the current absorbed by the balancing hardware associated with \( k^{\text{th}} \) cell. Battery pack life optimization is achieved by controlling each individual cell while taming state and parameters heterogeneity - due to either/both manufacturing defects or/and non uniform operating conditions. The formulated optimal control problem (OCP) will ultimately implement SOC balancing along with State of Health (SOH)-aware balancing by tackling the cell-to-cell heterogeneity. The optimal control is multi-objective in nature to face the conflicting objectives of minimum time of charge \( t_{f_k} \) under minimum degradation by optimizing the current profiles, \( I_{0} \) and \( I_{B_k} \) for \( k = 1, \ldots, N_{\text{cell}} \).\(^2\)

Cells in the module are modeled via coupled nonlinear PDEs, ODEs, and DAEs describing the electrochemical, thermal, and aging dynamics [25]. Single particle model (SPM) is employed to model the electrochemical dynamics, a lumped two-state thermal dynamical 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 a fast dynamics, the electrochemical dynamics is the semi-slow dynamics, and aging dynamics is the slow dynamics [17]. The nonlinear and multi-time scale nature of the cell dynamics are retained in the formulation and solution of the multi-objective 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. Noting the fact that aging dynamics include SEI layer growth and solvent concentration, where the former is the low-dimensional slow variable while the latter is the high-dimensional one. The computational burden imposed by running high fidelity dynamics at different time scale, has led to the design of a surrogate model to approximate the high-dimensional slow dynamics (solvent concentrations) as functions of cell currents and ambient temperatures.

To solve the optimization problem, the direct collocation approach [26] is utilized to transcribe the OCP to a nonlinear programming (NLP) problem [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 solutions are discussed using the the Karush-Kuhn-Tucker (KKT) conditions (first-order necessary conditions). The OCP is formulated under two different schemes: (a) \textit{same-charging-time} (OCP-SCT) and (b) \textit{different-charging-time} (OCP-DCT). To confirm the soundness of the proposed OCP-SCT and OCP-DCT schemes, simulation studies are carried out on an illustrative example: a battery module with two series-connected cells, each equipped with a active balancing hardware. The performance and robustness of the proposed schemes is shown under perturbation of parameters in terms initial SOC and initial SOH imbalances (through variation in the initial SEI layer growth state).

This paper 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) characterizes the time scale difference of the LIB dynamics, 3) provides ample simulation scenarios of the two optimization schemes for an effective and exhaustive comparison of the two and 4) 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

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

\(^2\)High C-rate currents would charge the battery faster at the expense of faster growth of Solid Electrolyte Interphase layer (SEI), causing capacity and power fade.
in the pursuit of a novel life-extension optimization charging strategy that combines advanced optimization algorithms over multi-scale high fidelity models using active balancing hardware setup.

C. Outline

The organization of the paper is as follows. **Section II** lists the notations used in the paper. **Section III** presents the battery module model. **Section IV** describes the problem statement. **Section V** formulates the proposed optimal control methodology. **Section VI** presents the simulation results. **Section VII** presents discussion and conclusion.

II. NOTATIONS

The following notations are used in the paper.

- Given a real \( n \)-dimensional vector \( x \) with initial and final values \( x(t_0) \) and \( x(t_f) \) (\( t_0 \) and \( t_f \) are the initial and final times), \( \Delta x = \frac{|x(t_f) - x(t_0)|}{x(t_0)} \times 100\% \) is the percentage deviation of \( x \) with respect to its initial value.
- Given the continuously differentiable function \( f(x) \), \( \nabla f(x) \) is the gradient of \( f(x) \) with respect to \( x \).
- The subscript \( j \in [n; p] \) stands for the cell domain (e.g. \( n \) = anode and \( p \) = cathode).
- The subscript \( i \) refers to the discretization grid position when converting PDEs to ODEs via Finite Difference Method (FDM) in solid electrodes and SEI layer spatial dimensions.
- The superscript \( k \) represents the cell position within the series-connected module.

III. BATTERY MODULE MODEL

This section presents the model for 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.

A. Cell Electrochemical Model

We use the SPM to model the electrochemical dynamics that 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 given by the Fick’s law

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

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

\[
\left. \frac{\partial c_{s,j}}{\partial r} \right|_{r=0} = 0
\]

\[
\left. \frac{\partial c_{s,j}}{\partial r} \right|_{r=R_{s,j}} = \frac{\pm I_{cell}}{D_{s,j}(T) a_{s,j} A L_j F} + g_{s,j}, \quad (2)
\]

where \( g_{s,j} \) is a general function of \( c_{surf}^{s,j}, c_{sol}^{s,j}, T_c, I_{cell} \), and \( L_{sei} \). 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 \( D_{s,j} \) and the reaction rate constant \( k_j \), follow an Arrhenius relationship with temperature given by

\[
\varphi(T_c) = \varphi_{ref} \exp \left[ \frac{E_a}{R_g} \left( \frac{1}{T_{c,ref}} - \frac{1}{T_c} \right) \right] \quad (3)
\]

with \( \varphi \) to be either \( D_{s,j} \) or \( k_j \).

The surface overpotentials of each electrode, \( \eta_j \) for \( j \in [n, p] \), is obtained from the Butler–Volmer kinetic equation describing the rate of intercalation and deintercalation of lithium ions as

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

where the exchange current density \( i_{0,j} \) is given by

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

From the concentration and potential distribution in the electrodes, the cell voltage \( V_{cell} \) can be calculated as

\[
V_{cell} = U_p(c_{surf}^{s,n}) + \eta_p(c_{surf}^{s,p}, c_{avg}^{s,p}, T_c, I_{cell}) - U_n(c_{surf}^{s,n}) - \eta_n(c_{surf}^{s,n}, c_{avg}^{s,n}, T_c, I_{cell}) - I_{cell} \left( R_l + R_{el} + R_{sei} \right) \quad (6)
\]

Fig. 1. Battery module with \( N_{cell} \) series-connected cells, where each cell is connected to a balancing hardware. The current variable \( I_0 \) refers to the module current, variables \( I_{cell1}, \ldots I_{cellN_{cell}} \) are the current magnitude through the cells, variables \( I_{B1}, \ldots I_{B_{N_{cell}}} \) are the balancing currents, and time variables \( t_{f1}, \ldots t_{f_{N_{cell}}} \) are the charging time associated with each cell.
in which the ohmic resistance includes the lumped contact resistance $R_l$, electrolyte resistance $R_{el}$, and SEI layer resistance $R_{sei}$, where the last two ones are given by

$$R_{sei} = \frac{L_{sei}}{a_s,n AL_n \kappa_{sei}},$$

$$R_{el} = \frac{1}{2A} \left[ \frac{L_n}{\kappa_{ei,n}} + \frac{2L_s}{\kappa_{ei,s}} + \frac{L_p}{\kappa_{ei,p}} \right],$$

where $\kappa_{ei,j}$ is a function of $c_{ei}^{avg}$ and $\epsilon_{ei,j}$. 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].

We calculate the bulk SOC of each electrode as

$$SOC_{bulk} = \frac{c_{ei,bulk} - \theta_0^j}{\theta_{100}^j - \theta_0^j} \quad j \in [n, p]$$

(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 paper, $SOC_{bulk}$ is used as battery cell SOC in the optimization algorithm design since cathode is the limiting electrode.

**B. Cell Thermal Model**

Thermal dynamics are modeled using the lumped parameter two-state thermal model

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

$$C_s \frac{dT_s}{dt} = T_{amb} - T_s - \frac{T_s - T_c}{R_c}$$

(9)

where $T_c$ and $T_s$ are the core and surface temperature of each cell. 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 anode SEI layer growth as a function of solvent reduction kinetics and diffusion dynamics to predict cell capacity loss and power fade. The solvent concentration available for reduction reaction at the anode surface is modeled by

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

(10)

with the boundary conditions

$$-D_{solv}(T) \frac{\partial c_{solv}}{\partial r} \bigg|_{r=R_{ei,n}} + \frac{dL_{sei}}{dt} c_{surf} \bigg|_{r=R_{ei,n}+L_{sei}} = \frac{i_s}{F}$$

(11)

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

$$\frac{dL_{sei}}{dt} = \frac{i_s M_{sei}}{2F \rho_{sei}}$$

(12)

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

$$i_s = -2Fk_f c_{surf}^2 e_{solv}^{surf} \exp \left( -\frac{-\beta_f}{R_g T_c} (\Phi_{s,n} - R_{sei} I_{cell} - U_s) \right).$$

(13)

The capacity loss is modeled by integrating side reaction current across the anode active surface area 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 cell-level state-space form can be derived. Note that DAEs are related to the Butler-Volmer equation used to calculate the overpotentials.

**Solid Phase Diffusion**: the state-space representation of the dynamics for each electrode is presented by

$$\dot{c}_{s,j} = \alpha_{s,j} A_{s,j} c_{s,j} + \beta_{s,j} B_{s,j} \left[I_{cell} - g_{s,j} \right],$$

(15)

where $c_{s,j} = [c_{s,j,1}, \ldots, c_{s,j,N_{r,j}}]^T \in \mathbb{R}^{N_{r,j}}$ with $c_{s,j,N_{r,j}} = c_{surf}$, $B_{s,j} = \left[0, \ldots, \left(2 + \frac{2}{N_{r,j} - 1}\right)\right]^T \in \mathbb{R}^{N_{r,j}}$,

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

(16)

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

(17)

and

$$g_{s,j}(c_{surf}, c_{solv}, I_{cell}, L_{cell}) = \begin{cases}
a_{s,n} L_n A_i & \text{if } j = n \\
0 & \text{if } j = p
\end{cases}$$

(18)

with $\Delta r_j = \frac{R_{ei,n}}{N_{r,j} - 1}$ and $N_{r,j}$ the number of radial discretization grids in SPM.

**SEI Layer Growth**: the ODEs for SEI layer growth and capacity loss are given by

$$\dot{L}_{sei} = \beta_{sei} g_{s,n} \quad \text{and} \quad \dot{Q} = \frac{\dot{L}_{sei}}{\rho_{sei}} = a_{s,n} L_n A_i$$

(19)

with $\beta_{sei} = \frac{-M_{sei}}{2F \rho_{sei} a_{s,n} L_n A_i}$.

**Solvent Diffusion**: the state-space form of the solvent diffusion dynamics is given by
where \( t \) whose dimension is dependent on the number of discretization points of the solvent diffusion PDE.

Thus, integrating the solvent diffusion dynamics (10) with the downstream and upstream cells, \( k \) with the number of SEI layer discretization points.

The difference in temporal scales in the cell dynamics is the cause of long - at time, prohibitive - simulation times which 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 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.

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]

\[
t_{\text{ter}} = \frac{R_{\text{cell}}^2}{\dot{\phi}}, \quad t_{\text{elec}} = \frac{R_{\text{cell}}^2}{D_{\text{solv}}}, \quad t_{\text{ag}} = \frac{R_{\text{cell}}^2}{D_{\text{solv}}},
\]

where \( t_{\text{ter}}, t_{\text{elec}}, \) and \( t_{\text{ag}} \) are the time scales of the thermal, electrochemical, and aging dynamics, respectively. Incorporating parameter values from the literature [17], [31], [32] shows that \( t_{\text{ag}} \) is in the order of \( 10^8 \) sec while \( t_{\text{ter}} \) and \( t_{\text{elec}} \) are in the orders of \( 10 - 100 \) sec and \( 10^3 \) sec, implying that the cell model is a three-time scale system in which \( t_{\text{ter}} < t_{\text{elec}} \ll t_{\text{ag}} \).

The state and parameter heterogeneity due to manufacturing defects and not uniform operating conditions are the cause of exacerbated aging of a battery pack when compared to a battery cell. The overall thermal and aging effects of the cells on the modules can be captured if the heat transfer between cells is properly modeled. The thermal interconnection between adjacent cells in the battery module is provided via the thermal resistance term \( R_{\text{m}} \) among a cell \( k \) with the downstream and upstream cells, \( k+1 \) and \( k+1 \), respectively. This results in the surface temperature dynamics to be modified as follows [33]

\[
C_s \frac{dT_{s_k}}{dt} = \frac{T_{\text{amb}} - T_{s_k}}{R_s} - \frac{T_{s_k} - T_{s_{k+1}}}{R_m} + \frac{T_{s_{k+1}} - T_{s_{k-1}}}{R_m}.
\]

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

A convenient shorthand notation for module-level dynamics with \( N_{\text{cell}} \) series-connected cells is as follows

\[
\begin{align*}
\dot{\epsilon}_{\text{solv}} & = \min_{\epsilon_{\text{solv}}} \| \dot{L}_{\text{sei}}^{hf} - L_{\text{sei}}^{hf}(\epsilon_{\text{solv}}) \|,
\end{align*}
\]

where \( L_{\text{sei}}^{hf} \) is the SEI layer thickness from the SPM inclusive of the solvent diffusion model (20), whereas \( L_{\text{sei}}^{hf}(\epsilon_{\text{solv}}) \) is the SEI layer thickness when constant solvent diffusion is used. Once optimal values of \( \epsilon_{\text{solv}}^* \) are found for different \( I_{\text{cell}} \) and \( T_{\text{amb}} \), \( \epsilon_{\text{solv}}^* \) -other polynomials \( G(I_{\text{cell}}, T_{\text{amb}}) \) are fitted to the optimal points \( \epsilon_{\text{solv}}^* \).

F. State-space representation: module-level

The state and parameter heterogeneity due to manufacturing defects and not uniform operating conditions are the cause of exacerbated aging of a battery pack when compared to a battery cell. The overall thermal and aging effects of the cells on the modules can be captured if the heat transfer between cells is properly modeled. The thermal interconnection between adjacent cells in the battery module is provided via the thermal resistance term \( R_{\text{m}} \) among a cell \( k \) with the downstream and upstream cells, \( k+1 \) and \( k+1 \), respectively. This results in the surface temperature dynamics to be modified as follows [33]

\[
C_s \frac{dT_{s_k}}{dt} = \frac{T_{\text{amb}} - T_{s_k}}{R_s} - \frac{T_{s_k} - T_{s_{k+1}}}{R_m} + \frac{T_{s_{k+1}} - T_{s_{k-1}}}{R_m}.
\]

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

A convenient shorthand notation for module-level dynamics with \( N_{\text{cell}} \) series-connected cells is as follows

\[
\begin{align*}
\epsilon_{s,j}^{\text{mod}} & = \epsilon_{s,j}^{\text{mod}} + B_{s,j}^{\text{mod}} u - G_{s,j}^{\text{mod}},
\end{align*}
\]

where \( \epsilon_{s,j}^{\text{mod}} \) is the SEI layer thickness from the SPM inclusive of the solvent diffusion model (20), whereas \( L_{\text{sei}}^{hf}(\epsilon_{\text{solv}}) \) is the SEI layer thickness when constant solvent diffusion is used. Once optimal values of \( \epsilon_{\text{solv}}^* \) are found for different \( I_{\text{cell}} \) and \( T_{\text{amb}} \), \( \epsilon_{\text{solv}}^* \) -other polynomials \( G(I_{\text{cell}}, T_{\text{amb}}) \) are fitted to the optimal points \( \epsilon_{\text{solv}}^* \).

\[
\begin{align*}
\epsilon_{solv} & = G_{solv}^\text{mod}.
\end{align*}
\]
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

\[
z(t) = \begin{bmatrix} c_{s,j}^\text{mod} \\ T^\text{mod} \\ \beta^\text{mod} \\ g^\text{mod} \\ c_{s,j}^\text{set} \end{bmatrix}^T.
\]

Note that the right hand side of the solvent diffusion dynamics, \( G \text{mod}_{\text{sol}} \), 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 \( A_{\text{mod}}^\text{solv} \), \( B_{\text{mod}}^\text{solv} \), and \( G^\text{mod}_{\text{sol}} \) vary between cells due to cell heterogeneity of design parameters, non-uniform aging, temperature distribution.

Upon the creation of the surrogate model to replace the solvent diffusion dynamics, presented in the previous subsection, the module state vector used in the proposed optimal control design is given by \(^3\)

\[
x(t) = \begin{bmatrix} c_{s,j}^\text{mod} \\ T^\text{mod} \\ \beta^\text{mod} \\ g^\text{mod} \\ c_{s,j}^\text{set} \end{bmatrix}^T.
\]

IV. OPTIMAL CONTROL PROBLEM FORMULATION

In this section, we formulate a multi-objective optimal control framework for fast charging and minimum degradation of a battery module with \( N_{\text{cell}} \) series-connected imbalanced cells as shown in Fig. 3. In this configuration, the module capacity coincides with the weakest cell in the string.

\(^3\)Note that \( c_{s,j}^\text{set} \) included in \( z(t) \) is now excluded from the system-level state vector \( x(t) \).

When dealing with battery modules accounting for the intrinsic heterogeneity among the cells in terms of charge, temperature and \( \text{SOH} \) is key. In particular, in this work battery health is defined both in terms of \( Q \) and \( R_{\text{sei}} \), both dependent on \( L_{\text{sei}} \) as seen from (7) and (19). To model cells suffering from \( \text{SOH} \) imbalances, selection of different initial conditions for \( L_{\text{sei}} \) is made.

An OCP is formulated for two different schemes OCP-SCT and OCP-DCT. The former assumes that all cells are charged simultaneously, irrespective of their non homogeneous initial states, whereas the latter assumes different times of charging of the cell to reflect the non uniform initial states the cells are at. In particular, OCP-DCT is aimed at providing a charge strategy that extends the battery life and provides more flexibility against heterogeneity among the cells, i.e., \( \text{SOC} \) and \( \text{SOH} \) imbalances. From Fig. 3, \( I_0 = I_{\text{cell}} + I_{\text{B}} \) for \( k = 1, \ldots, N_{\text{cell}} \) from which one can define the vector

\[
I_{\text{cell}} = [I_{\text{cell}} \ldots I_{\text{cell}}]^{N_{\text{cell}}} = [I_0 - I_{\text{B}} \ldots I_0 - I_{\text{B}_{\text{cell}}}]^{T}.
\]

During charging, the module current \( I_0 \in \mathbb{R} \) and the vector of balancing currents \( I_B = [I_{\text{B}} \ldots I_{\text{B}_{\text{cell}}}]^{T} \in \mathbb{R}^{N_{\text{cell}}} \) are unknown and must be optimally planned; the number of degrees of freedom (DoF) is \( N_{\text{DoF}} = N_{\text{cell}} + 1 \). Each cell is connected in parallel to an active balancing circuitry whose current \( I_B \) is determined by the proposed optimal controller.

### Table I

**Module-level matrices and state vectors.**

| Component | Matrix/Vector | Description |
|-----------|---------------|-------------|
| Solid phase diffusion: | \( A_{\text{mod}}^\text{solv} = \begin{bmatrix} (\alpha_{s,j} A_{s,j})_1 \\ \vdots \\ (\alpha_{s,j} A_{s,j})_{N_{\text{cell}}} \end{bmatrix} \) | Coefficient matrices for solid phase diffusion dynamics. |
| Thermal: | \( A_{\text{therm}}^\text{mod} = \begin{bmatrix} \frac{1}{R_t C_t} \\ \frac{1}{R_t C_t} \end{bmatrix} \) | Coefficient matrices for thermal dynamics. |
| State vectors: | \( \begin{bmatrix} c_{s,j}^\text{mod} \\ T^\text{mod} \\ \beta^\text{mod} \\ g^\text{mod} \\ c_{s,j}^\text{set} \end{bmatrix} \) | State vectors for module level. |
| Aging: | \( G^\text{mod}_{\text{set}} = \begin{bmatrix} \beta_{\text{set}}(g_{s,n})_1 \\ \vdots \\ \beta_{\text{set}}(g_{s,n})_{N_{\text{cell}}} \end{bmatrix} \) | Coefficient matrices for aging dynamics. |
| | \( G^\text{mod} = \begin{bmatrix} [g_{s,n}]_1 \\ \vdots \\ [g_{s,n}]_{N_{\text{cell}}} \end{bmatrix} \) | Coefficient matrices for aging dynamics. |
The following OCP for the OCP-DCT scheme is formulated as:

$$X^* = \underset{X \in \mathbb{R}^{N_{opt}}}{{\text{argmin}}} \alpha \beta_1 h(t_f) + \beta_2 \sum_{k=1}^{N_{cell}} \sum_{l=1}^{L_{sei,k}} \beta_3 g_2(\hat{L}_{sei,k})$$

(28)

where the vector of optimization variables $X$ is comprised of the vector of final times of charging $t_f = [t_{f1} \ldots t_{fN_{cell}}]^T \in \mathbb{R}^{N_{cell}}$, the system state $x(t) \in \mathbb{R}^{N_s}$, the module current $I_0$, and the balancing current vector $I_B$:

$$X = [t_f, x(t), I_0(t), I_B(t)]^T \in \mathbb{R}^{N_{opt}}.$$  

(29)

The number of optimization variables is $N_{opt} = N_s + 2N_{cell} + 1$, the number of states is $N_s = N_{cell} (4 + 2(N_r - 1))$ for a given $N_r$ and the state vector defined in (26), and the continuously differentiable functions $g_1$, $g_2$, and $h$ are defined as

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

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

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

(30)

Note that $g_1(L_{sei}), g_2(\hat{L}_{sei}),$ and $h(t_f)$ are operators that return the average of SEI layer thicknesses, the average of their rates, and the average of charging times, respectively.

Thus, the OCP (28) along with the definitions (30) forms a \textit{min-mean optimization problem}. The positive scalars $\beta_1 [s^{-1}], \beta_2 [\text{sm}^{-1}],$ and $\beta_3 [\text{sm}^{-1}]$ are optimization weights corresponding to the charging time and SEI layer growth objectives, respectively, 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 $0 \leq \alpha \leq 1$ is a trade-off 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 $k = 1, \ldots, N_{cell}$. To establish safety metrics, module and balancing currents, voltages, core and surface temperatures, and solid concentrations of all cells are enforced to lie within some physical bounds for $k = 1, \ldots, N_{cell}$

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

$$V_{cell_{min}} \leq V_{cell_k}(t) \leq V_{cell_{max}},$$

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

(31)

$$0 \leq \theta_{0, c_{s,j_{max}}} \leq \theta_{100\% c_{s,j_{max}}}, \quad j \in \{n, p\}.$$  

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

$$L_{sei_{k}}(t_0) = L_{sei_{ik}k}, \quad Q_{k}(t_0) = Q_{0_k},$$

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

(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:

$$0 \leq t_{f_k} \leq t_{f_{max}}.$$  

(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

1) The final times of charging are the same for all cells

Fig. 3. Battery module with $N_{cell}$ series-connected cells, where each cell is connected to a balancing hardware. The proposed optimal controller realized through Nonlinear Programming using Direct Collocation optimizes the variables annotated in blue for fast charging and minimized degradation.
from which one can consider \( t_f \in \mathbb{R} \).
2) The number of optimization variables reduces to \( N_{opt} = N_s + N_{cell} + 2 \); hence
\[
\mathbf{X} = [t_f, \mathbf{x}(t), I_0(t), \mathbf{I}_B(t)]^T \in \mathbb{R}^{N_{opt}},
\]
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 \( 0 \leq t_f \leq t_{f_{\text{max}}} \) in which the charging time of all cells is the same.

The next section is devoted to solve the OCP (28) subject to dynamic constraints (24) and the operating constraints (31)-(35).

V. OPTIMAL CONTROL ALGORITHM

In this paper, 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 a NLP problem [27] by approximating all elements of the unknown vector \( \mathbf{X} \) 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 break points (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 can be approximated as
\[
X_p(t) = \sum_{q=1}^{N_{FP_0}} B_{p,q} \omega_{p,q} \quad \text{for} \quad p = 1, \ldots, N_{opt},
\]
where \( B_{p,q} \) and \( \omega_{p,q} \) are the \( q^{th} \) B-spline and free parameter of the \( p^{th} \) optimization variable, and \( N_{FP_0} = N_p(d_p - s_p) + s_p \) is the number of free parameters for the \( p^{th} \) optimization variable with \( N_p = N_{BP} - 1 \) as the number of polynomial segments [34]. By parameterizing all of the system trajectories \( t_f, \mathbf{x}(t), I_0(t), \) and \( \mathbf{I}_B(t) \) (\( t_f \) is scalar in case of OCP-SCT), the total number of free variables is calculated as
\[
N_{FP}^r = N_{FP_0} + (N_{FP_{B_p}} + N_{FP_{I_B}}) N_{cell} + N_{FP_{I_0}},
\]
where \( N_{FP_{B_p}}, N_{FP_{I_B}}, N_{FP_{I_0}}, \) and \( N_{FP_r} \) are the numbers of free parameters for each state, balancing and module currents, and charging times, respectively; they are design parameters to be selected by users.

With this approximation in hand, the original OCP (28) can be transcribed to the NLP problem of the general mathematical form
\[
P^* = \arg\min_P J(P)
\]
s.t.
\[
g_{P_1}(P) = 0, \quad g_{P_2}(P) \leq 0, \quad P_{\text{min}} \leq P \leq P_{\text{max}},
\]
where \( P = [\omega_{p,q}] \in \mathbb{R}^{N_{BP} \times 2} \) is the finite set of free parameters; and \( J \in \mathbb{R} \), and \( g_{P_1} \in \mathbb{R}^{m_1} \) and \( g_{P_2} \in \mathbb{R}^{m_2} \) 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 \( P \).

The Lagrangian function \( \mathcal{L} : \mathbb{R}^{N_{FP} \times 2} \times \mathbb{R}^{m_1} \times \mathbb{R}^{m_2} \rightarrow \mathbb{R} \) associated with the NLP problem (39) is defined as
\[
\mathcal{L}(P, \mu_1, \mu_2) = J(P) + \mu_1^T g_{P_1}(P) + \mu_2^T g_{P_2}(P) \tag{40}
\]
with \( \mu_1 \in \mathbb{R}^{m_1} \) and \( \mu_2 \in \mathbb{R}^{m_2} \). The Karush-Kuhn-Tucker (KKT) optimality conditions [35] associated with (40) are
\[
\nabla \mathcal{L} = \nabla J(P^*) + \sum_{r=1}^{m_1} \mu^*_r \nabla g_{P_1}(P^*) \\
+ \sum_{r=1}^{m_2} \mu^*_r \nabla g_{P_2}(P^*) = 0 \quad \text{(Stationarity)},
\]
\[
g_{P_1}(P^*) = 0 \quad \text{for} \quad r = 1, \ldots, m_1
\]
\[
g_{P_2}(P^*) \leq 0 \quad \text{for} \quad r = 1, \ldots, m_2 \quad \text{(Primal feasibility)},
\]
\[
\mu^*_r \geq 0 \quad \text{for} \quad r = 1, \ldots, m_2 \quad \text{(Dual feasibility)},
\]
\[
\sum_{r=1}^{m_2} \mu^*_r g_{P_2}(P^*) = 0 \quad \text{(Complementary slackness)},
\]
where Conditions (41)-(44) are called Stationarity, Primal feasibility, Dual feasibility, and Complementary slackness, respectively; and \( \mu^*_r \) for \( r = 1, \ldots, m_1 \) and \( \mu^*_r \) 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 \( (\mu^*_1, \mu^*_2) \) such that the KKT conditions (41)-(44) hold, then a solution \( P^* \) is a local optimum for the NLP problem (39). It should be pointed out that when \( P^* \) and \( (\mu^*_1, \mu^*_2) \) are any primal dual optimal points with zero duality gap (strong duality), then any pair of \( (P^*, (\mu^*_1, \mu^*_2)) \) satisfies the KKT conditions (41)-(44) [35].

Under the direct collocation approach, the cost and constraints are applied to the optimization variables \( t_f, \mathbf{x}(t), I_0(t), \) and \( \mathbf{I}_B(t) \) (\( t_f \) is scalar in case of OCP-SCT) at collocation points (CPs). In this paper, we determine the CPs based on the Gaussian quadrature formula (GQF) using which the BPs do not coincide with the CPs. 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 dynamics and operating constraints, and the cost are written symbolically. This formulation provides symbolic differentiation of the OCP, which in turn, results in remarkable improvement in convergence time and solving reliability.
Remark 1: In view of (38), the number of free parameters reduces to $N_{FP} = N_{FP_c} + N_{FP_{c,j}} + N_{FP_{c,j}}$, when 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

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                     |

Fig. 4. Open-circuit potentials of NMC cathode/graphite anode cell.

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. 3).

A. Initialization and set up

The battery considered in this paper is a cylindrical 18650, 2-Ah lithium-ion nickel–manganese–cobalt (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, $\theta_{surf} = \theta_{surf}^{c,j} / \theta_{surf}^{s,j}$, is illustrated in Fig. 4. Throughout the simulations, we assume that there is an initial SOC imbalance among the cells ($SOC_1(0) \neq SOC_2(0)$) 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 $I_{B_{max}} = -6$ A, $I_{B_{min}} = -16$ A, $I_{B_{max}} = 0$ A, $I_{B_{sat}} = -12$ A, $SOC_{target} = 0.8$, $V_{cell_{min}} = 2.5$ V, $V_{cell_{max}} = 4.2$ V, $t_{f_{max}} = 2000$ s, $T_{b_{max}} = 5^\circ$C, and $T_{b_{max}} = 45^\circ$C 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 $L_{sei1}(0) = 5 \times 10^{-9}$ m (this is the typical SEI later thickness observed for a fresh cell), $Q_k(0) = 2$ Ah, and $T_{c_k}(0) = T_{c_k}(0) = T_{amb}$. The numbers of discretization points are set to $N_{f} = N_{i} = 44$. Thus, the number of states used in the OCP is $N_{s} = 44$. Given the nominal capacity $Q_{nom} = Q_k(0) = 2$ Ah, the selected bounds for module and balancing currents result in the c-rate to lie between 3C and 8C. For the balanced charging-degradation scenario, the optimization weights and the trade-off coefficient 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 sampling current 2 A) and ambient temperatures are chosen to be $[15, 25, 35]$ °C. $5^{th}$-order polynomials are fitted to the optimal points $e_{sol}^{c}$ 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.

B. Initial SOC mismatch with different ambient temperatures

At an ambient temperature of $T_{amb} = 25^\circ$C, we first solve OCP-SCT and OCP-DCT schemes when the initial SOCs are set to $SOC(0) = [0.2, 0.4]$. From Fig. 5, under OCP-SCT scheme, both cells are charged simultaneously while their voltages lie within $V_{cell_{min}} = 2.5$ V and $V_{cell_{max}} = 4.2$ 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 $I_{B1}$ results in the higher $I_{cell1}$).

In contrast, charging times are different for Cell 1 and Cell 2 when using the OCP-DCT scheme as demonstrated in Figs. 6. As expected, Cell 2 (with 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 $t_{f2}$, (i) $SOC_2$ is kept constant until Cell 1 reaches $SOC_{target}$ at $t_{f1}$, (ii) the cell current is absorbed by the power units implementing the active balancing circuitry, leading to $I_0 = I_{B2}$ over $t_{f2} \leq t \leq t_{f1}$, (iii) $V_{cell_{2}}$ drops at $t_{f2}$ and remains constant over $t_{f2} \leq t \leq t_{f1}$, (iv) the core and surface temperatures of Cell 2 start decreasing at $t_{f2}$, and (v) the rates of $L_{sei2}$ and $Q_2$ slow down after $t_{f2}$. These figures also show that the OCP-DCT scheme reduces degradation gradient between cells (i.e., $L_{sei2}(t_{f2}) - L_{sei1}(t_{f1})$) at all different ambient temperatures.

To compare the results of OCP-SCT and OCP-DCT, Table III lists quantitative comparisons between two schemes at different ambient temperatures ($T_{amb} = [15, 25, 35]^\circ$C). Referring to this table, under each OCP-SCT or OCP-DCT, when $T_{amb}$ increases, the following trends are inferred: (i) the SEI layer thickness variation of both cells increases, (ii) the capacity lose variation of either cell increases, and (iii) the charging time of either cell increases. According to this table, OCP-DCT decreases $max(\Delta L_{sei1}, \Delta L_{sei2})$ by 73%, 40%, and 40% over OCP-SCT when $T_{amb}$ is set to $15^\circ$C, $25^\circ$C, and $35^\circ$C, respectively; this, in turn, leads to
max(ΔQ₁, ΔQ₂) to be decreased by 72%, 40%, and 35%, respectively when OCP-DCT is used. In terms of charging time, however, OCP-DCT increases max(t₁, t₂) by 40%, 28%, and 42% over OCP-SCT when Tₐmb 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 in the presence of random initial SOC and SOH imbalances, this section is devoted to perform multiple simulations at different ambient tempera-
TABLE III

PERFORMANCE COMPARISON BETWEEN THE OCP-SCT AND OCP-DCT SCHEMES WITH INITIAL SOC MISMATCH $SOC_1(0) = 0.2$ AND $SOC_2(0) = 0.4$ AT DIFFERENT AMBIENT TEMPERATURES $T_{amb} = [15, 25, 35]^\circ C$. THE BEST VALUE OF EACH METRIC IS SHOWN IN BOLD.

| $T_{amb}$ | Scheme   | $(\Delta L_{sei1}^*, \Delta L_{sei2}^*)$% | $(\Delta Q_1, \Delta Q_2)$% | $(t_{f1}^*, t_{f2}^*)$ s |
|----------|----------|------------------------------------------|----------------------------|--------------------------|
| 15°C     | OCP-SCT  | (0.079, 0.021)                           | (20, 5.5)$\times 10^{-4}$  | (406, 406)               |
|          | OCP-DCT  | (0.021, 0.021)                           | (5.43, 5.41)$\times 10^{-5}$ | (571, 386)               |
| 25°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°C     | OCP-SCT  | (60, 47)                                 | (0.15, 0.12)                | (471, 471)               |
|          | OCP-DCT  | (38, 29)                                 | (0.098, 0.073)              | (671, 458)               |

Numerical results show that under OCP-SCT scheme, $\max_{k_{N_{sim}}} (\Delta L_{sei1}^*, \Delta L_{sei2}^*) = \{0.08, 9.62, 57.15\}$% and $\max (t_{f1}^*, t_{f2}^*) = \{510, 596, 717\}$ s for $T_{amb} = [15, 25, 35]^\circ C$, where $k_{N_{sim}} = 1, \ldots, 20$ is the $k_{N_{sim}}$ simulation. On the other hand, with OCP-DCT scheme and under different ambient temperatures, $\max_{k_{N_{sim}}} (\Delta L_{sei1}^*, \Delta L_{sei2}^*) = \{0.04, 6.22, 38.64\}$% and $\max (t_{f1}^*, t_{f2}^*) = \{587, 646, 738\}$ s. These findings are in agreement with our observations in Table III, showing that the optimization under 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, \( N_{sim} = 20 \) simulations are run for each ambient temperature for both control scheme where in each simulation, initial \( L_{sei} \) values are drawn from a uniform distribution over the interval \([4, 6] \times 10^{-9} \text{m}\) to represent the SOH imbalance at the beginning of the battery life. From Fig. 8, results reveal that with OCP-SCT scheme, \( \max \left( \Delta L_{sei1}^*, \Delta L_{sei2}^* \right) = [0.09, 14.94, 77.10]\% \) and \( \max(t_f^*) = [498, 547, 783] \text{s} \), and under OCP-DCT scheme, \( \max \left( \Delta L_{sei1}^*, \Delta L_{sei2}^* \right) = [0.06, 9.98, 57.87]\% \) and \( \max \left( t_{f1}^*, t_{f2}^* \right) = [546, 620, 815] \text{s} \) for all \( T_{amb} = [15, 25, 35] \text{°C} \). These results are in line with what we found from the case of initial SOC imbalance, showing that OCP-DCT scheme is able to mitigate the variation of SEI layer thickness at the cost of higher charging time no matter what ambient temperature is used. In comparison with the case of initial SOC imbalance, results also reveal that the simulations with initial SOH imbalance leads to the battery module with higher variation of SEI layer thickness at any ambient temperature used.

D. Pareto fronts: effect of trade-off coefficient

Recall that the optimization trade-off coefficient was picked to be \( \alpha = 0.5 \) in the previous sections to have balanced charging-degradation 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, \( \alpha \) 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. 8 shows that the maximum of SEI layer thickness variations of the cells reduces as \( \alpha \) 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 \( \alpha \) goes from 1 to 0, at \( T_{amb} = [15, 25, 35] \text{°C} \), (i) under OCP-SCT, \( \max \left( \Delta L_{sei1}^*, \Delta L_{sei2}^* \right) \) decreases by 72%, 38%, and 23%, and \( \max(t_f^*) \) increases by 66%, 49%, and 75%, respectively; and (ii) under OCP-DCT, \( \max \left( \Delta L_{sei1}^*, \Delta L_{sei2}^* \right) \) decreases by 71%, 26%, and 27%, and \( \max \left( t_{f1}^*, t_{f2}^* \right) \) increases by 97%, 46%, and 53%, respectively. Once again, the Pareto fronts support our previously-claimed observations, showing that 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 constant current (CC) charging profiles. Given that research efforts are underway to enable extreme fast charging, wherein the battery pack must be charged to 80% of its capacity in 10-15 minutes [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. The performance of the series-connected cells under the four charging profiles [3C, OCP-DCT, OCP-SCT, 8C] are evaluated at an ambient temperature of 25°C in terms of (a) charging time for the first cycle, and (b) capacity loss at the end of 300 cycles. In this case, the capacity loss for a cell is defined as the percentage change in its capacity at the end of 300 cycles, with respect to the nominal capacity, given by \( \Delta Q_{loss} = \frac{Q_{nom} - Q_{cyc}}{Q_{nom}} \times 100\% \). This study intends to demonstrate the health savings each charging strategy offers, in terms of retained capacity, over multiple charging cycles. As observed in Table III, the OCP-DCT profile always provides a better performance than the OCP-SCT.

\( ^4 \) The CC-CV charging protocol is used in laboratory testing, whereas only CC - or its variants- is used for in-vehicle charging.
in terms of minimized degradation. Hence, the comparison shown here is to prove that OCP-SCT and OCP-DCT outperform the CC profiles. To evaluate the performance, the module of two cells in series with initial SOC imbalance \( SOC(0) = [0.2, 0.4] \) is simulated when subjected to the charging profiles of 3C, OCP-DCT, OCP-SCT, and 8C, separately, for a duration of 300 charging cycles. Note that one cycle is composed of the cells being charged from their initial SOC to the final SOC of 0.8.

In Fig. 10(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, respectively. It is noticed that the charging time reduces as the C-rate increases from 3C to 8C, and the charging time for OCP-SCT and OCP-DCT profile sits between the two extremes. As expected, the amount of degradation has the opposite trend wherein as the C-rate increases, the observed capacity loss is higher. However, interestingly, the capacity loss observed for the OCP-SCT and OCP-DCT profile is much lower than 8C and slightly lower than 3C. 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 trade-off 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. 10(b) for all charging profiles. Cell 2 has a higher initial SOC, and hence its charging time for the OCP-DCT profile is much faster because the scheme allows for different charging times of the cells to account for heterogeneous initial conditions, whereas the charging time of Cell 2 is same as that of Cell 1 for the OPT-SCT profile. The results validate that the OCP-DCT and OCP-SCT profile outperform the standard CC profiles by providing a balanced trade-off between fast charging and minimum degradation. Note that these results are simulated for 300 charging cycles, however, each cycle only consists of a SOC window from 0.2 or 0.4 to 0.8 (depending on 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 paper formulated a multi-objective 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 under 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 against heterogeneity among the cells in terms of obtaining a more uniform degradation among the cells, hence leading to a longer utilization and higher reliability 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 paper, which consists of using the direct collocation approach to transcribe the OCP into a nonlinear programming (NLP) problem by parameterization of the system states and inputs, for a series-connected module, will remain the same.

B. Discussion: Impact of our work

The adoption of an 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. In a series-connected module, the capacity of the module is defined by the weakest (most aged) cell. Heterogeneity among cells, if not tamed, 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 paper positions itself as an easily deployable method for targeted applications.

VIII. Acknowledgements

The authors are grateful to LG Chem (now LG ES) for its financial support, and they would like to thank Dr. Won Tae Joe and Dr. Yohwan Choi for providing invaluable guidance for this work.

REFERENCES

[1] S. M. Lukic, J. Cao, R. C. Bansal, F. Rodriguez, and A. Emadi, “Energy storage systems for automotive applications,” IEEE Transactions on Industrial Electronics, vol. 55, pp. 2258–2267, June 2008.

[2] A. Hoke, A. Brissette, D. Maksimovic, A. Pratt, and K. Smith, “Electric vehicle charge optimization including effects of lithium-ion battery degradation,” in 2011 IEEE Vehicle Power and Propulsion Conference, pp. 1–8, Sep. 2011.

[3] H. Farzin, M. Fothu-Firuzabad, and M. Moemi-Aghtaie, “A practical scheme to involve degradation cost of lithium-ion batteries in vehicle-to-grid applications,” IEEE Transactions on Sustainable Energy, vol. 7, pp. 1730–1738, Oct 2016.

[4] Y. Du, J. Wu, S. Li, C. Long, and S. Onori, “Hierarchical coordination of two-time scale microgrids with supply-demand imbalance,” IEEE Transactions on Smart Grid, vol. 11, pp. 5726–3736, Sep. 2020.

[5] Y. Liu, C. Hsieh, and Y. Luo, “Search for an optimal five-step charging pattern for li-ion batteries using consecutive orthogonal arrays,” IEEE Transactions on Energy Conversion, vol. 26, pp. 654–661, June 2011.
Fig. 10. Charging time vs 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.

### Acronyms

- **BP** Break point
- **CCCV** Constant-current constant-voltage
- **CP** Collocation point
- **DAE** Differential algebraic equation
- **DOD** Depth of discharge
- **DCT** Different charging time
- **ECM** Equivalent circuit models
- **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

---

[6] K. Liu, K. Li, Z. Yang, C. Zhang, and J. Deng, “An advanced lithium-ion battery optimal charging strategy based on a coupled thermoelectric model,” *Electrochimica Acta*, vol. 225, pp. 330 – 344, 2017.

[7] C. Zou, C. Manzie, and D. Nesić, “Model predictive control for lithium-ion battery optimal charging,” *IEEE/ASME Transactions on Mechatronics*, vol. 23, no. 2, pp. 947–957, 2018.

[8] B. Suthar, V. Ramadesigan, P. W. C. Northrop, B. Gopaluni, S. Santhanagopalan, R. D. Braatz, and V. R. Subramanian, “Optimal control and state estimation of lithium-ion batteries using reformulated models,” in *2013 American Control Conference*, pp. 5350–5355, June 2013.

[9] X. Hu, S. Li, H. Peng, and F. Sun, “Charging time and loss optimization for linnmc and lifepo4 batteries based on equivalent circuit models,” *Journal of Power Sources*, vol. 239, pp. 449 – 457, 2013.

[10] F. Lam, A. Allam, W. T. Joe, and S. Choi, Y.and Onori, “Offline multiobjective optimization for fast charging and reduced degradation in lithium-ion battery cells using electrochemical dynamics,” *IEEE Control Systems Letters*, vol. 5, pp. 2066–2071, 2021.

[11] M. Pathak, D. Sonawane, S. Santhanagopalan, R. D. Braatz, and V. R. Subramanian, “(invited) analyzing and minimizing capacity fade through optimal model-based control - theory and experimental validation,” *ECS Transactions*, vol. 75, pp. 51–75, Jan 2017.

[12] A. Allam, S. Onori, S. Marelli, and C. Taborelli, “Battery health management system for automotive applications: a retroactivity-based aging propagation study,” in *American Control Conference (ACC)*, 2015, pp. 703–716. IEEE, 2015.

[13] D. Beck, P. Dechent, M. Junker, D. Sauer, and M. Dubarry, “Inhomogeneities and cell-to-cell variations in lithium-ion batteries, a review,” *Energies*, vol. 14, 2021.

[14] A. Barré, B. Deguilhem, S. Grolleau, M. Gérard, F. Suard, and D. Riu, “A review on lithium-ion battery ageing mechanisms and estimations for automotive applications,” *Journal of Power Sources*, vol. 241, pp. 680 – 689, 2013.

[15] J. Vetter, P. Novák, M. Wagner, C. Veit, K.-C. Möller, J. Besenhard, M. Winter, M. Wohlfahrt-Mehrens, C. Vogler, and A. Hammouche, “Ageing mechanisms in lithium-ion batteries,” *Journal of Power Sources*, vol. 147, no. 1, pp. 269 – 281, 2005.

[16] F. Todeschini, S. Onori, G. Rizzoni, and A. Cordoba-Arenas, “An experimentally validated capacity degradation model for li-ion batteries inhevps applications,” in *8th IFAC Symposium on Fault Detection, Supervision and Safety of Technical Processes*, vol. 45, pp. 456–461, 2012.

[17] A. Allam and S. Onori, “Exploring the dependence of cell aging dynamics on thermal gradient in battery modules: A pde-based time scale separation approach,” in *2019 18th European Control Conference (ECC)*, pp. 2380–2385, 2019.

[18] J. Gallardo-Lozano, E. Romero-Cadaval, M. I. Milanes-Montero, and M. A. Guerrero-Martínez, “Battery equalization active methods,” *Journal of Power Sources*, vol. 246, pp. 934–949, 2014.

[19] M. M. U. Rehman, F. Zhang, M. Evzelman, R. Zane, and D. Maksimovic, “Control of a series-input, parallel-output cell balancing system for electric vehicle battery packs,” in *2015 IEEE 16th Workshop on Control and Modeling for Power Electronics (COMPEL)*, pp. 1–7, 2015.

[20] Y. Li and Y. Han, “A module-integrated distributed battery energy storage and management system,” *IEEE Transactions on Power Electronics*, vol. 31, no. 12, pp. 8260–8270, 2016.

[21] D. J. Docimo and H. Fathy, “Multivariable state feedback control as a foundation for lithium-ion battery pack charge and capacity balancing,” *Journal of The Electrochemical Society*, vol. 164, 2017.

[22] F. Altaf, B. Egardt, and L. Johannesson Mårdh, “Load management of hybrid electric vehicle battery packs,” in *2015 IEEE 18th Workshop on Control and Modeling for Power Electronics (COMPEL)*, pp. 1–7, 2017.

[23] D. J. Docimo and H. K. Fathy, “Analysis and control of charge and temperature imbalance within a lithium-ion battery pack,” *IEEE
TABLE IV
NOMENCLATURE.

| Symbol | Description                                      |
|-------|--------------------------------------------------|
| c_{n,j} | Concentration in solid phase [mol/m^3]          |
| c_{e}  | Concentration in electrolyte phase [mol/m^3]   |
| c_{solc} | Solvent concentration [mol/m^3]               |
| T_{c}  | Cell core temperature [K]                        |
| T_{s}  | Cell surface temperature [K]                     |
| Q      | Cell capacity [Ah]                              |
| I_{cell} | Cell current [A]                          |
| I_{ext} | SEI layer thickness [m]                          |
| η_{j}  | Overpotential [V]                                |
| u_{o,j} | Exchange Current Density [A/m^2]                |
| U_{j}  | Open circuit potential (electrode) [V]           |
| i_{x}  | Side reaction current density [A/m^3]            |
| D_{x,j} | Solid phase diffusion [m^3/s]                    |
| R_{x,j} | Particle radius [m]                              |
| a_{x,j} | Specific interfaceal surface area [m^-1]       |
| A      | Cell cross sectional area [m^2]                  |
| L_{j}  | Domain thickness [m]                             |
| F      | Faraday’s constant [C/mol]                      |
| c_{exo,j} | Maximum electrode concentration [mol/m^3]  |
| c_{el,f} | Effective electrolyte conductivity [S/m]      |
| k_{f}  | Reaction rate constant [m^2/s-mol^0.5]          |
| R_{SEI} | SEI layer resistance [Ω]                        |
| R_{g}  | Universal gas constant [J/mol-K]                |
| D_{adv} | Solvent diffusion in SEI layer [m^3/s]          |
| R_{sei} | Electrolyte resistance [Ω]                      |
| ε_{sei} | SEI layer porosity [Kg/m^3]                     |
| n_{sei} | SEI layer ionic conductivity [S/m]              |
| c_{solc} | Molar mass of SEI layer [kg/mol]               |
| C_{se} | Heat Capacity of cell surface [J/K]             |
| C_{c}  | Heat Capacity of cell core [J/K]                |
| R_{m}  | Conductive resistance - cell/surface [K/W]      |
| T_{amb} | Ambient temperature [K]                         |
| N_{r,j} | Number of radial discretization points         |
| c_{ego} | Average electrolyte concentration [mol/m^3]    |
| c_{eso,f} | Surface concentration in solid phase [mol/m^3]  |
| c_{solc,f} | Surface solvent concentration [mol/m^3]         |
| r_{f}  | Solvent Reduction rate constant [mol^-2s^-1]    |
| c_{solc} | Optimal solvent concentration [mol/m^3]        |
| I_{cell} | Cell current [A]                              |
| V_{cell} | Cell voltage [V]                              |
| T_{ref,c} | Reference core temperature [K]                |
| ε_{el} | Electrolyte porosity                           |
| Φ_{s,n} | Anode surface 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_{BP} | Number of break points                          |
| Q_{nom} | Nominal capacity [Ah]                           |
| α,β    | Optimization parameters                         |
| s_{p}  | Smoothness degree                               |
| p      | Free parameter set                              |
| μ_{p,q} | B-Spline                                        |
| φ      | Thermal diffusivity                              |
| R_{cell} | Radius of a cylindrical 18650 cell [m]       |

Transactions on Control Systems Technology, vol. 27, pp. 1622–1635, July 2019.
[24] A. Pozzi, M. Zambelli, A. Ferrara, and D. M. Raimondo, “Balancing-aware charging strategy for series-connected lithium-ion cells: A nonlinear model predictive control approach,” IEEE Transactions on Control Systems Technology, vol. 28, no. 5, pp. 1862–1877, 2020.
[25] T. Weaver, A. Allam, and S. Onori, “A novel lithium-ion battery pack modeling framework - series-connected case study,” in 2020 American Control Conference (ACC), pp. 365–372, July 2020.
[26] C. R. Hargraves and S. W. Paris, “Direct trajectory optimization using nonlinear programming and collocation,” AIAA J. Guidance, vol. 10, no. 4, p. 338–342, 1987.
[27] A. Rao, “A survey of numerical methods for optimal control,” Advances in the Astronautical Sciences, vol. 135, no. 1, p. 497–528, 2009.
[28] A. Wachter, L. Biegler, Y. Lang, and A. Raghunathan, “Ipopt: An interior point algorithm for large-scale nonlinear optimization,” Advances in the Astronautical Sciences, 2002.
[29] V. Azimi, A. Allam, W. T. Joe, Y. Choi, and S. Onori, “Fast charging-minimum degradation optimal control of series-connected battery modules with dc/dc bypass converters,” in 2021 American Control Conference (ACC), pp. 231–236, 2021.
[30] X. Lin, H. E. Perez, S. Mohan, J. B. Siegel, A. G. Stefanopoulou, Y. Ding, and M. P. Castanier, “A lumped-parameter electro-thermal model for cylindrical batteries,” Journal of Power Sources, vol. 257, pp. 1–11, 2014.
[31] S. Al Hallaj, H. Maleki, J. Hong, and J. Selman, “Thermal modeling and design considerations of lithium-ion batteries,” Journal of Power Sources, vol. 83, no. 1, pp. 1–8, 1999.
[32] H. Maleki, S. A. Hallaj, J. R. Selman, R. B. Dinwiddie, and H. Wang, “Thermal properties of lithium-ion battery and components,” Journal of The Electrochemical Society, vol. 146, pp. 947–954, mar 1999.
[33] A. Allam and S. Onori, “Characterization of aging propagation in lithium-ion cells based on an electrochemical model,” in 2016 American Control Conference (ACC), pp. 3113–3118, 2016.
[34] R. Bhattacharya, “Optragen: A matlab toolbox for optimal trajectory generation,” in Proceedings of the 45th IEEE Conference on Decision and Control, pp. 6832–6836, Dec 2006.
[35] S. Boyd and L. Vandenberghe, Convex optimization. Cambridge university press, 2004.
[36] T. R. Tanin, E. J. Dufek, M. Evans, C. Dickerson, A. N. Jansen,
B. J. Polzin, A. R. Dunlop, S. E. Trask, R. Jackman, I. Bloom, et al., “Extreme fast charge challenges for lithium-ion battery: variability and positive electrode issues,” Journal of The Electrochemical Society, vol. 166, no. 10, p. A1926, 2019.