BETTER BATTERIES

THROUGH ELECTROCHEMISTRY

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atteries are everywhere: in our smar phones, laptops, electric vehicles (EVs), and electric grids. Energy storage is a critical enabling technology for enhancing energy sustainability. Although battery materials science has seen rapid advances, the systems are underutilized and conservatively designed. Consumers purchase batteries with 20-50% excess energy capacity, leading to added weight, volume, and upfront cost. Intelligent battery control can lead to faster charge times, increased energy and power capacity, as well as a longer life. The key to realizing such advanced battery management systems is electrochemistry and controls—a fusion of modern control theory and electrochemical models that allows batteries to operate safely at their physical limits.

This article introduces key concepts in ElectroChemical-based Control (ECC) systems for batteries, and highlights the fundamentals of battery electrochemistry, state-of-charge/state-of-health estimation, and constrained control.

BATTERY FUNDAMENTALS

Jumping Frog Legs: A Brief History of the First Battery

talian physicist Alessandro Volta invented the first battery cell in 1800. The so-called voltaic pile consisted of two metals in series, zinc and copper, coupled by a sulphuric acid electrolyte. Volta's inspiration came from experiments performed by his colleague Luigi Galvani who was interested in the interaction between electricity and biological nervous systems. During his experiments, Galvani discovered that a dead frog's legs would kick to life when in contact with two dissimilar metals. Volta reasoned that the different metals caused this behavior, and demonstrated this to be true with his voltaic pile.

Principles of Operation

battery, put simply, converts between chemical and electrical energy through oxidation-reduction reactions. As shown by the zinc-copper Galvanic cell in Figure 1, it consists of two dissimilar metals (electrodes) immersed in an electrolyte. The cathode and anode materials are selected to have a large electrochemical potential between each other. This provides the desired electrochemical energy storage property. The electrodes are electrically isolated from each other via a separator. Hence, electrons are forced through an external circuit, powering a connected device, while cations flow between the electrodes within the electrolyte.

Electrode and electrolyte materials are selected for their voltage, charge capacity, weight, cost, manufacturability, etc. For example, lithium-ion cells are attractive in mobile applications because lithium is the lightest (6.94 g/mol) and most electropositive (-3.01V vs. standard hydrogen electrode) metal in the periodic table. Lead acid cells feature heavier electrodes (Pb and PbO₂), yet provide high surge currents at cost effec-

tive prices. Lithium-air batteries feature cathodes that couple electrochemically with atmospheric oxygen, thus producing energy densities that rival gasoline fuel.

In battery energy management, we are interested in maximizing performance and longevity. This requires a detailed understanding of the underlying electrochemistry. However, the electrochemical variables are not directly measurable. At best, one can measure voltage, current, and temperature only. Consequently, modeling and control are necessary to extract the full potential from batteries.

State-of-Charge (SOC) Estimation

OC indicates the remaining charge, analogous to a fuel tank level indicator. Unlike fuel tanks, SOC is not measurable—it is estimated by combining models and measurements.

To motivate the particular challenges of SOC estimation consider **Figure 2**, which provides the relationship between voltage and SOC at different C-rates (*see sidebar below "What is C-rate?"*) for a LiFePO₄ cell. In principle, one may measure voltage and invert the nonlinear relationship shown in Fig. 2. Two challenges are immediately visible. First, voltage shifts with C-rate, thus complicating inversion. Second, function inversion is highly sensitive to measured voltage errors, since the slope is nearly zero in the 5%–95% SOC range. In addition, this relationship varies with temperature, age, and cell chemistry. Accurate models and estimation theory are needed to address this problem, especially in highly dynamic and safety critical environments such as electric vehicles.

State-of-Health (SOH) Estimation

The two most common SOH metrics are charge capacity fade and power capacity fade. Charge capacity fade indicates how charge capacity has decreased relative to its nameplate value (e.g., a 2 Ah cell may hold 1.6 Ah after two years of use). Power capacity fade indi-

WHAT IS C-RATE?

C-rate is a normalized measure of electric current that enables comparisons between different sized batteries. Mathematically, the C-rate is defined as the ratio of current, *I*, in Amperes (A) to a cell's nominal capacity, *Q*, in Ampere-hours (Ah). For example, if a battery has a nominal capacity of 2.5 Ah, then C-rates of 2C, 1C, and C/2 correspond to 5A, 2.5 A, and 1.25 A, respectively. Note that C-rate has dimensions of [A] / [Ah] = [1/h].

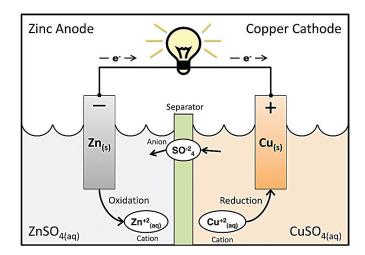


FIGURE 1 Zinc-copper Galvanic cell demonstrating the principles of operation for an electrochemical cell.

cates how power capacity has decreased relative to its nameplate value (e.g., a fresh cell may provide 360W of power for 10 seconds, but only 300W after two years of use). Gradual changes in SOH metrics can be related to changes in a mathematical model's parameters. A rich body of literature on parameter identification is readily available to address SOH estimation. Several interesting challenges arise within the context of electrochemical models, including derivations of parametric models, nonlinear parameter identifiability, and persistency of excitation.

Controlled Charging/Discharging

In current applications, additional capacity is added to mitigate cell imbalance, capacity/power fade, thermal effects, and estimation errors. This leads to larger, heavier, and more costly batteries than required. ECC alleviates oversizing by safely operating batteries near their physical limits. Today, operation is defined by voltage, current, and temperature limits—all measurable variables. Battery degradation, however, is more closely related to limits on the immeasurable electrochemical states, such as overpotentials and surface concentrations. Consequently, we seek a paradigm-shifting architecture that expands the operating envelope by constraining internal electrochemical states instead of voltage, current, and temperature, as seen in Figure 3. This combines SOC/SOH estimation with control algorithms to form a comprehensive ECC battery management system.

MATHEMATICAL MODELING

Equivalent Circuit vs. Electrochemical Models

athematical battery models generally fall into two categories: equivalent circuit models (ECM) and electrochemical models (EChem); see **Figure 4**. ECMs predict the input-output behavior of cells via electric circuits. The simplest ECM, shown in Fig. 4(a), considers a battery as a nonlinear voltage source in series with an internal resistor. This is written in state-space form as

$$S\dot{O}C(t) = -\frac{1}{O}I(t)$$

$$V(t) = OCV(SOC(t)) - RI(t)$$

where I(t) is current, Q is charge capacity, $OCV(\cdot)$ is the open circuit voltage function, R is internal resistance, and V(t) is voltage. These models become more complex and accurate by considering additional circuit components (e.g. RC pairs) and more parameterizations. Although ECMs are intuitive to formulate, they are insufficient for controlling the electrochemical states. Many researchers have recently focused solely on EChem models.

EChem models capture the spatiotemporal dynamics of lithium-ion concentration, electric potential, and intercalation kinetics. Most models in the battery controls literature are derived from the Doyle-Fuller-Newman (DFN) model $_2$, which is based upon porous electrode and concentrated solutions theory. Fig. 4b shows a cross section of the layers described in Fig. 1. At full charge most of the lithium exists within the anode solid phase particles, typically lithiated carbon Li_xC_6 , that are idealized as spherically symmetric. During discharge, lithium diffuses from the interior to the surface of these spherical particles. At the surface an electrochemical reaction separates lithium into a positive lithium ion and electron.

$$\text{Li}_x \text{C}_6 \rightleftharpoons \text{C}_6 + \chi \text{Li}^+ + \chi \text{e}^-$$

Next, the lithium ion migrates from the anode, through the separator, and into the cathode. Since the separator is an electrical insulator, the corresponding electron travels through an external circuit, powering the connected device. The lithium ion and electron meet at the cathode particles' surface, typically a lithium metal oxide LiMO $_2$, and undergo the reverse electrochemical reaction.

$$\text{Li}_{1-x}\text{MO}_2 + \chi \text{Li}^+ + \chi \text{e}^- \rightleftharpoons \text{LiMO}_2$$

The produced lithium atom then diffuses into the interior of the cathode's spherical particles. This entire process is reversible by applying sufficient potential across the current collectors – rendering an electrochemical storage device. In addition to lithium migration, this model captures the spatial-temporal dynamics of internal potentials, electrolyte current, and current density between the solid and electrolyte phases.

Although EChem models predict battery operation over broad conditions, they are mathematically complex. **Table 1** summarizes the main equations, including partial differential equations (PDEs), ordinary differential equations (ODEs) in space, ODEs in time, and nonlinear algebraic constraints. This complexity prohibits estimator and control design. As a result, there is a focus on reduced-order models that facilitate control design while predicting the dynamics of interest.

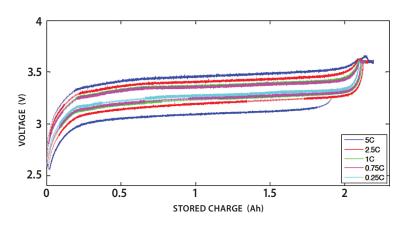


FIGURE 2
Relationship between terminal voltage, storage charge, and C-rate for a LiFePO4 cell.

Reduced-Order Models

rapidly growing body of literature is establishing a spectrum of EChem models that achieve varying balances of mathematical simplicity and accuracy. The most fundamental reduced EChem model is the single particle model (SPM). The SPM idealizes each electrode as a single aggregate spherical particle. This model results if one assumes the electrolyte Li concentration $c_e(x,t)$ from is constant in space and time. This assumption works well for small currents, yet produces errors at large C-rates. Mathematically, the model consists of two diffusion PDEs governing each electrode's concentration dynamics,

$$\frac{\partial c_s^-}{\partial t}(r,t) = D_s^- \left[\frac{2}{r} \frac{\partial c_s^-}{\partial r}(r,t) + \frac{\partial^2 c_s^-}{\partial r^2}(r,t) \right]$$
 5

$$\frac{\partial c_s^+}{\partial t}(r,t) = D_s^+ \left[\frac{2}{r} \frac{\partial c_s^+}{\partial r}(r,t) + \frac{\partial^2 c_s^+}{\partial r^2}(r,t) \right]$$
 6

where input current enters as Neumann boundary conditions

$$\frac{\partial c_s^-}{\partial t}(0,t) = 0, \qquad \frac{\partial c_s^-}{\partial t}(R_s^-,t) = \frac{I(t)}{D_s^- F a^- A L^-}$$

$$\frac{\partial c_s^+}{\partial t}(0,t) = 0, \qquad \frac{\partial c_s^+}{\partial t}(R_s^+,t) = \frac{-I(t)}{D_s^+ F a^+ A L^+}$$

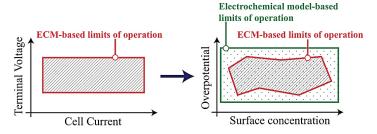
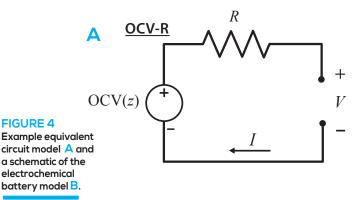


FIGURE 3 Current battery management systems regulate operation by limiting measurable quantities (e.g. voltage and current). An ECC approach expands the operating regime by regulating the immeasurable electrochemical states within safe limits.



The boundary conditions at $r = R_s^+$ and $r = R_s^-$ signify that flux is proportional to input current I(t). Output voltage is given by a nonlinear function of the state values at the boundary $c_{ss}^+(t)$, $c_{ss}^-(t)$ and the input current I(t) as follows

$$V(t) = \frac{RT}{\alpha F} \sinh^{-1} \left[\frac{I(t)}{2\alpha^{+}AL^{+}i_{0}^{+}(c_{ss}^{+}(t))} \right] - \frac{RT}{\alpha F} \sinh^{-1} \left[\frac{I(t)}{2\alpha^{-}AL^{-}i_{0}^{-}(c_{ss}^{-}(t))} \right]$$

$$+ U^{+}(c_{ss}^{+}(t)) - U^{-}(c_{ss}^{-}(t)) + R_{f}I(t)$$
9

where the $i_0^j(\cdot)$ is the exchange current density and $c_{\infty}^j(t) = c_{\infty}^j(R_s^j,t)$ is the surface concentration for electrode $j \in \{+,-\}$. The functions $U^{j}(\cdot)$ are the equilibrium potentials of each electrode material, given the surface concentration.

The SPM reduces the DFN model to two linear state equations and a nonlinear output mapping. This model is amenable to control/observer design, however its predictive capability is limited to low C-rates. Other researchers have developed higher-fidelity reduced EChem models via a swath of numerical methods₄₋₈. Nonetheless, these models are not always oriented towards controller/observer synthesis, thus motivating further research.

A critical property for state estimation is observability. The DFN model is not completely observable (in the linear sense) from voltage, current, and temperature measurements. Several heuristics have been successfully applied to render complete observability, which are associated with reduced-order modeling 3,4,7. However, exploitation of nonlinear observability or PDE observability remains an open opportunity.

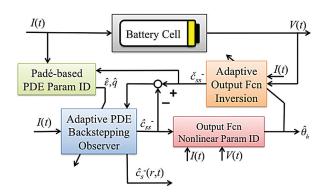
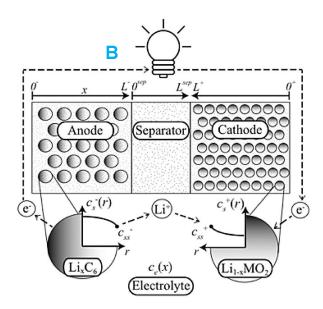


FIGURE 5 Block diagram of the adaptive observer composed of the backstepping state observer (blue), PDE parameter identifier (green), output function parameter identifier (red), and adaptive output function inversion (orange). The observer furnishes estimates of SOC (i.e. $\hat{c}_{5}(r,t)$) and SOH (i.e. $\hat{\varepsilon}, \hat{q}, \hat{\theta}_h$) given measurements of I(t) and V(t), only.



STATE-OF-CHARGE/STATE-OF HEALTH ESTIMATION

esearch on battery SOC/SOH estimation has experienced considerable growth, and can be categorized under ECM or EChem model-based algorithms.

The first category considers estimators based upon ECMs. For example, the seminal work by Plett 9. applies an extended Kalman filter to simultaneously identify the states and parameters of an ECM. The key advantage of ECMs is their simplicity. However, they are unable to predict relevant electrochemical states and parameters. The second category considers electrochemical models. Although these models can predict internal states, their complex mathematical structure prohibits controller/observer design. These approaches employ model reduction with estimation. Some of the first studies within this category used the SPM in combination with an extended Kalman filter 10. Another approach uses residue grouping for model reduction and Kalman filters for observers 11. The authors of reference 6 apply approximations to the electrolyte and solid concentration dynamics to perform SOC estimation. More recently, simultaneous SOC and SOH estimation was performed on a SPM using PDE-theoretic techniques 3. Simultaneous SOC and SOH estimation using electrochemical models is in infancy, and represents a rich problem for dynamic systems and control researchers.

Adaptive PDE Observer for Single Particle Models

e present a simultaneous SOC/SOH estimation algorithm using adaptive PDE observer designs based upon a SPM. The SOC and SOH estimation problems can be cast mathematically as state and parameter estimation problems, respectively. That is, SOC can be defined in terms of the anode solid concentration $c_s^-(r,t)$ and SOH can be defined in terms of electrochemical parameters, such as moles of cyclable lithium n_{Ii} and electrolyte resistance R_f .

Figure 5 summarizes the complete algorithm. Although the SPM is a relatively simple electrochemical model, it

FIGURE 4

Description	Equation	Eqn.	
Solid phase Li concentration	$\frac{\partial c_s^{\pm}}{\partial t}(x,r,t) = \frac{1}{r^2} \frac{\partial}{\partial r} \left[D_s^{\pm} r^2 \frac{\partial c_s^{\pm}}{\partial r}(x,r,t) \right]$	(10)	
Electrolyte Li concentration	$\varepsilon_s \frac{\partial c_e}{\partial t}(x,t) = \frac{\partial}{\partial x} \left[\varepsilon_e D_e \frac{\partial c_e}{\partial x}(x,t) + \frac{1 - t_c^0}{F} i_e^{\pm}(x,t) \right]$	(11)	
Solid potential	$\frac{\partial \phi_s^{\pm}}{\partial x}(x,t) = \frac{i_e^{\pm}(x,t) - I(t)}{\sigma^{\pm}} $ (12)		
Electrolyte potential	$\frac{\partial \phi_e}{\partial x}(x,t) = -\frac{i\frac{1}{e}(x,t)}{\kappa} + \frac{2RT(1-t_c^0)}{F} \left(1 + \frac{d\ln f_c^0}{d\ln c_e}\right) \frac{\partial \ln c_e}{\partial x}(x,t)$	(13)	
Electrolyte ionic current	$\frac{\partial i_e^2}{\partial x}(x,t) = a_s F j_n^{\pm}(x,t)$	(14)	
Butler-Volmer kinetics	$j_n^{\pm}(x,t) = \frac{2}{F}i_0^{\pm}(x,t)\sinh\left[\frac{\alpha F}{RT}\eta^{\pm}(x,t)\right]$		
Temperature	$\rho c_p \frac{dT}{dt}(t) = h[T^{\infty}(t) - T(t)] + I(t)V(t)$ $-\int_{0^{-}}^{0^{+}} a_s F j_n(x, t) \Delta T(x, t) dx$	(16)	

TABLE 1 Main equations for the electrochemical model.

contains several notable challenges. These include (1) the PDE dynamics, (2) the nonlinearity imposed by estimating states and parameters together, and (3) the output function's nonlinear relationship with respect to both states and parameters. We take a cascaded design approach to address these issues. That is, we synthesize identification algorithms for uncertain parameters in the PDE state equation and output function. These estimates are then applied to a backstepping PDE state observer algorithm, using the certainty equivalence principle₃.

To demonstrate, we consider a vehicle-like charge/discharge cycle generated from two concatenated urban dynamometer driving schedule (UDDS) drive cycles. This signal is highly transient with large C-rate magnitudes, producing a sufficiently rich signal for parameter estimation. **Figure 6** portrays the state and parameter estimates using data generated from the DFN model summarized in Table 1. The state estimates are represented by bulk SOC and surface concentration, which converge to their true values. The PDE parameter estimates \hat{e}, \hat{q} and output function parameter estimates $\hat{n}_{Li}\,\hat{R}_f$ (normalized to one in Fig. 6) also converge near their true values. Similar results are achievable for various other initial conditions and charge/discharge cycles. The relative complexity of combined SOC/SOH estimation for the simplest of electrochemical models highlights the problem richness. Open research opportunities include algorithms based on higher-fidelity models, nonlinear estimation, novel sensing, and experimental verification.

TABLE 2 Electrochemical states to be constrained within upper/lower limits.

Variable	Definition	Constraint
I(t)	Current	Power electronics limits
$c_s^{\pm}(x,r,t)$	Li concentration in solid	Material saturation/depletion
$\frac{\partial c_s^{\pm}}{\partial r}(x,r,t)$	Li concentration gradient in solid	Diffusion-induced stress/strain
$c_e(x,t)$	Li concentration in electrolyte	Material saturation/depletion
T(t)	Temperature	High/low temp. accelerate aging
$\eta_s(x,t)$	Side reaction overpotential	Li-plating, SEI-layer growth

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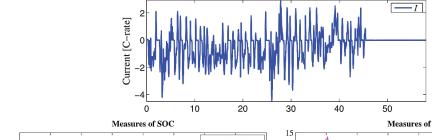
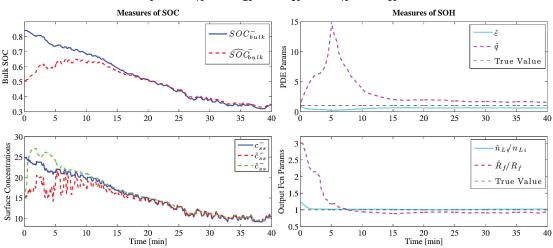


FIGURE 6 Evolution of state and parameter estimates for two concatenated UDDS charge/discharge cycles. The DFN model summarized by Table 1 provides the "measured" plant data. State and parameter estimates were initialized with incorrect values.



CONTROLLED CHARGING/DISCHARGING

attery packs are typically oversized and underutilized to ensure longevity and robust operation. Indeed, oversizing mitigates degradation mechanisms, such as lithium plating, lithium depletion/over-saturation, overheating, and stress fractures by reducing C-rates. However, oversizing can be overly conservative. In this section we discuss concepts for eliminating this conservatism. Namely, constrained optimal control methods, such as reference governors (RG), enable smaller-sized batteries whose electrochemical states satisfy safe operating constraints.

Constrained Control

nsuring safe operating constraints is a basic requirement for batteries. Mathematically, this can be abstracted as a constrained control problem for which RGs provide one promising solution. We seek to maintain operation subject to electrochemical state constraints. This protects the battery against catastrophic failure and maintains longevity, an issue underscored by the recent Boeing 787 Dreamliner battery failures₁₂. A list of relevant state constraints is provided in **Table 2**. These limits are associated with material saturation/depletion, mechanical stress, extreme temperatures, and harmful side reactions, such as lithium plating and solid/electrolyte interphase film growth.

A reference governor is an add-on device that guarantees state constraint satisfaction pointwise-in-time while tracking a desired reference input₁₃. In our "modified" reference governor (MRG) implementation, the applied current $I^r[k]$ and reference current are related according to

$$I[k+1] = \beta[k]I^r[k], \qquad \beta \in [0,1]$$
 17

where I[k] = I(t) for $\epsilon [k\Delta t, (k+1)\Delta t], k \epsilon Z$. The goal is to maximize β such that the state stays within an admissible region over some future time horizon,

$$\beta^*[k] = \max\{\beta \in [0,1]: x(t) \in O\}$$
 18

Variable x(t) represents the electrochemical model state at time t and θ is the set of initial conditions that maintain the state within the constraints listed in Table 2, over a future time horizon $\tau \in [t, t + T_s]$. See reference 14.

Figure 7 compares the standard constant current-constant voltage (CCCV) protocol, to an MRG that utilizes perfect estimates of the constrained states. CCCV applies 1C charging until voltage reaches a manufacturer-specified "maximum," 4.2V in this case. Next, CCCV regulates terminal voltage at the maximum voltage, 4.2V, while current diminishes toward zero. The value of 4.2V is selected to avoid lithium plating caused by overcharging. Mathematically, this corresponds to $\eta_s \ge 0$ in Fig. 7. Indeed, the side reaction overpotential remains positive, however it is conservative. Specifically, the side reaction overpotential can be regulated closer to zero. The MRG applies 1C charging subject to the constraint η_s ≥ 0 . In Fig. 7 the MRG maintains $\eta_s \geq 0$ despite voltage exceeding 4.2V. Moreover, the cell attains 95% SOC in 24min vs. 38min for CCCV. Note that CCCV reaches a final SOC of 96%, whereas the MRG achieves 100% SOC. Consequently, charging time is decreased by 37% and energy capacity is increased by 4%. Note that initial C-rates above 1C in the constant current region can further enhance performance beyond what is shown here.

One combines this MRG design with a state observer to form a complete output feedback ECC system (**Figure 8**). Several questions remain, such as robustness to estimation errors, forecasting reference current, optimal charge/discharge current trajectories, and experimental validation.

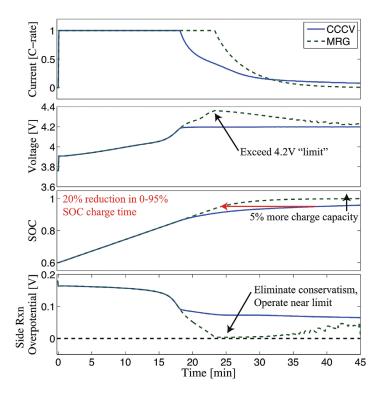


FIGURE 7 Comparison of CCCV and modified reference governor (MRG) charging. The MRG regulates near its limit, thereby achieving 95% SOC in 24min vs. 38min for CCCV, despite voltage exceeding 4.2V.

FUTURE OUTLOOK

atteries play a prominent role in developing technologies to ensure energy security, enhance sustainability, and lower greenhouse gases. However, today's reality is that batteries are expensive and conservatively designed. Advanced control systems that optimize battery performance and longevity are a key enabler for reducing costs and catalyzing deeper penetration into transportation fleets and electric power grids. Namely, promising solutions exist at the nexus of electrochemical modeling and advanced control theory. The dynamic systems and control community is uniquely positioned to play a significant role, as batteries provide a rich opportunity for advancements in fundamental control science and emerging energy application areas.

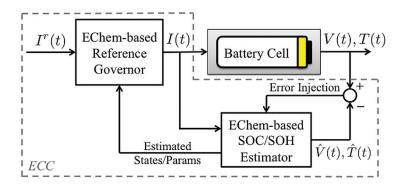


FIGURE 8 Block diagram of the ECC system comprised of a SOC/SOH estimator to determine the electrochemical states/parameters, and a reference governor to apply controlled charging/discharging.

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