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Perspective—Moving Next-Generation Phase-Field Models to BMS Applications: A Case Study that Confirms Professor Uzi Landau's Foresight

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Physics-based electrochemical models play a prominent role in the model-based analysis, virtual engineering, and Battery Management Systems (BMS) of lithium-ion and next-generation batteries. In this paper, we demonstrate the rich physics of phase-field models and convey their potential in BMS applications. Our phase-field model-based optimization framework predicts an impulse-like control profile to reduce capacity degradation. This work was partially inspired by the pulse-charging protocol proposed by Professor Landau in his 2006 work [B. K. Purushothaman and U. Landau, *J Electrochem Soc*, **153**(3), A533 (2006)]. An open-source framework is shared for predicting the (im)pulse protocol reported in this paper.
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Batteries serve as energy storage devices for a variety of applications, from consumer electronics to electric vehicles and renewable energy storage. Battery models can serve two primary purposes for all of these use cases. Prior to manufacturing the battery cell, battery models can be utilized to drive design by linking the performance requirements of a battery cell in a particular system to the electrode and material level design choices during the development phase.¹ After a battery has been implemented into the target system, battery models are developed to predict the physical state of a battery and track degradation dynamics to optimize performance, efficiency, and long-term health.² Battery Management Systems (BMS) sustain safe battery operating conditions, manage thermal loads, and perform fault detection during operation to detect scenarios that lead to undesired events.^{3,4}

Physics-based electrochemical models are essential tools that facilitate the simulation of electrochemical and transport phenomena inside the battery, enable battery parameter estimation, and inform control-based cycling protocols for lithium-ion (Li-ion) batteries.⁵ BMS implemented with physics-based models can also estimate the state-of-charge (SOC) and state-of-health (SOH) of the battery. Physics-based models consist of coupled differential-algebraic equations (DAEs) that can vary in complexity and simulation time based on the granularity of the underlying physics. These models, compared to empirical approaches, have greater accuracy and better adaptability for the simulation of rapid cycling protocols, new cell chemistries, and novel degradation mechanisms. With the adoption of batteries in the automotive and energy sectors, the accurate prediction of capacity degradation and remaining useful life (RUL)

of batteries has become critical. Various degradation mechanisms, such as solid electrolyte interphase (SEI) growth, particle cracking, and lithium plating contribute to the shortening of life and must be captured in order to mitigate operational concerns present in aged battery systems.⁶ BMS informed by electrochemical models are well-suited to incorporate degradation phenomenon into predictive models and give manufacturers and users meaningful inputs to assist in operating the batteries. Physics-based BMS are particularly useful in feedforward prediction of high-rate operation scenarios in electric vehicle operation, specifically with respect to fast charging and range estimates during towing, both of which serve to mitigate consumer range anxiety and drive domestic electric vehicle adoption. However, the computational costs associated with physics-based battery models can be prohibitive for both onboard estimation and control applications, as well as offline parameter identification and control optimization.² Therefore, model reformulation and reduced order models are often needed to implement advanced physics-based BMS methods and improve computational efficiency.^{5,7} Finally, the recent trends in data-driven modeling also make use of physics-based electrochemical modeling via physics-informed machine learning models that can aid in improving the computational footprint.⁸

In the realm of physics-based modeling, models of different complexity, time, and length scales have been developed to study batteries for BMS and systems engineering applications.^{5,9} The diffusion process within the cell electrodes has typically been modeled using Fick's laws of diffusion. Recently, the intercalation of lithium-ions in the electrode particle was posed as a phase change problem, where the area intercalated with lithium undergoes phase change compared to pristine material. Such phase change behavior is found in cathodes such as lithium iron phosphate (LFP) and anodes such as Graphite.¹⁰ With a refocus in the automotive sector on LFP: Graphite batteries, consideration of the phase change during intercalation can aid in both the virtual engineering domain during

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development, and the controls domain while in operation. Researchers have approached this phase change problem in terms of evolving core-shell problems^{11,12} and, more recently, using phase field models^{13,14} that may hold certain advantages over the traditional Fickian diffusion models.

Phase field models originated from diffuse-interface models developed by Cahn & Hilliard in the 1950s.¹⁵ They are based upon thermodynamics and characterized by a diffuse interface that introduces the phase-field variable to indicate the phase state of a material system.¹⁶ As the system evolves, the phase-field variable directly reflects the phase transition process which is derived according to principles of local equilibrium and free energy minimization, typically described by nonlinear partial differential equations (PDEs) and solved numerically. Phase-field models have been applied to batteries to gain deeper insights into the physics of lithiation,¹⁷ lithium plating (Li-plating),¹⁸ particle cracking,¹⁹ and dendritic growth.²⁰ Phase-field models are relevant for both the current and next generation of battery chemistries, but they need to be properly parametrized, formulated, and simulated for robust and convergent simulations. In addition, as shown in a recent paper, kinetics-driven interface growth requires simulation of Hamilton Jacobi equations.¹⁸

Electrochemical model-based control approaches remain at the forefront of efforts to improve the overall life and performance of Li-ion batteries. Battery design parameters such as electrode thickness and porosities are optimized using full battery models as well as reduced-order models.^{21,22} Optimum charging protocols are obtained to improve battery lifetime and reduce degradation.^{23–25} Pathak et al. developed a model-based optimal charging approach for a NMC based Li-ion pouch cell that resulted in a cycle life improvement of over 100%, compared to a standard CC-CV charging method.²⁶ Recent efforts have been focused on minimizing Li-plating via optimized charging protocols during regular cycling^{27,28} and extreme fast charging.²⁹ The model-based investigation on the effects of temperature and cell parameters on the performance of CC-CV and boost charging profiles has shown that under conditions that favor shorter diffusion timescales, performance is better with the boost charging profiles.³⁰ Finally, Landau et al.³¹ have analyzed constant current and pulse current profiles for Fickian diffusion (in a simulation mode, not optimal control mode) and argued that pulse current reduces the charging time and reduced concentration polarization-related losses. This work has been built upon by adding more physics and mechanisms subsequently.^{32–34}

This work is partially inspired by the pulse-charging protocol that was proposed by Professor Landau in his 2006 paper.³¹ In that work, empirically selected pulse charging profiles that show enhanced charging were modeled using Fick's law of diffusion. It was shown that using pulsed charging profiles avoided lithium saturation at the surface, enabling charging at higher rates. This was a model-based examination of empirical pulsed profiles used by experimentalists and not an effort to arrive at an optimal profile using Fick's law for diffusion in particles. In the present work, we have shown that, using phase field formulations in place of Fick's model actually predicts a pulsed profile as an optimal charging profile, thereby providing theoretical framework for empirically observed pulsed profiles during experiments. Thus, we have aimed to integrate the recent advances in phase-field models for batteries with the model-based control and optimization approaches from our previous works.^{23,35}

Current Status

Intercalation modeled with Fickian diffusion.—Traditionally, Fickian diffusion has been used to model the intercalation of Li-ions within the electrode particles.³⁶ Though the spherical domain is more appropriate for battery models, this paper reports models in cartesian coordinates (rectangular slab) for clarity and numerical ease. In this case, the electrode is assumed to be a planer rectangular electrode with symmetry at $x = 0$, while the other is in contact with electrolyte ($x = 1$). The cartesian results are applicable to spherical

particles as well. The galvanostatic charge for constant diffusivity for a fully discharged cathode particle can be written as:

$$\begin{aligned} t_d \frac{\partial c}{\partial t} &= -\frac{\partial N_x}{\partial x} = \frac{\partial^2 c}{\partial x^2}; N_x = -\frac{\partial c}{\partial x} \\ t &= 0; c(0, t) = 1 \\ x = 0, -\frac{\partial c}{\partial x} &= N_x = 0 \\ x = 1, -\frac{\partial c}{\partial x} &= N_x = \delta t_d \end{aligned} \quad [1]$$

In this model, c and x are dimensionless concentration and distance, t is dimensional time (s), and t_d is the diffusion time constant ($\frac{R^2}{D}$). N_x represents the flux of concentration. If we consider $c = 0$ as the fully delithiated (charged) state of the cathode, corresponding to a fully charged state of the battery cell (SOC = 100%), then $\delta = \frac{1}{3600}$ is the scaled current density for a C-rate of 1 C. Volume averaging of Eq. 1 gives $\frac{dc_{avg}}{dt} = -\delta$ showing mass conservation consistent with the Faraday's law. For a given current density and t_d , this model can predict concentration profiles at different times. Based on the Butler-Volmer or Marcus theory, one can then find the overpotential at $x = 1$ i.e. at the electrode and electrolyte interface. Assuming that there is no kinetic limitation, the potential of the electrode is given by $V = 3.42 - OCV(c_x)$ where c_x is the dimensionless concentration at $x = 1$. 3.42 V is the observed potential for LiFePO₄ at 50% SOC as reported in the Zeng and Bazant paper.¹³ Pseudo-two-dimensional (P2D) type models based on Newman's seminal work,³⁶ build on these models with different variations (nonlinear diffusivity) for the solid-phase diffusion. Note that to predict the voltage profile, one needs an Open-Circuit Voltage (OCV) fitting from the experimental data, or an algebraic representation such as the Multi-Species, Multi-Reaction model.^{37,38}

Cahn hilliard reaction model.—Zeng and Bazant¹³ presented a thermodynamically improved version of the intercalation model based on the Cahn-Hilliard Reaction (CHR) model. This model can be written (with some modifications for ease of simulation and analysis) as,

$$\begin{aligned} t_d \frac{\partial c}{\partial t} &= -\frac{\partial N_x}{\partial x}; N_x = -c(1-c) \frac{\partial \mu}{\partial x} \\ \mu &= \log\left(\frac{c}{1-c}\right) + \omega(1-2c) - \kappa \frac{\partial^2 c}{\partial x^2} \\ t &= 0; c(0, t) = 0.999 \\ x = 0, \frac{\partial c}{\partial x} &= 0; N_x = -c(1-c) \frac{\partial \mu}{\partial x} = 0 \\ x = 1, \frac{\partial c}{\partial x} &= 0; N_x = -c(1-c) \frac{\partial \mu}{\partial x} = \delta t_d \end{aligned} \quad [2]$$

Note that t_d is added in this paper compared to Zeng and Bazant's¹³ paper, and rectangular coordinates are used). N_x here again refers to the modified flux expression. μ is the chemical potential, whereas ω and κ are the enthalpy of mixing per site and the gradient energy penalty coefficient in dimensionless form respectively.¹³ The ω parameter may be regarded as a scaled molar enthalpy of the mixing parameter. It is a measure of the strength of interactions between intercalated ions and vacant sites in the active material. A negative value of ω indicates greater attraction between the intercalated ions and sites. This value thus predicts single-phase behavior throughout the Li stoichiometry range. A positive value of ω higher than a certain threshold ($\omega > 2$) leads to the formation of two intercalant phases. We have chosen two representative values denoting single phase and phase separating dynamics. These values correspond to the LFP system in this case. The theoretical maximum

for SOC is 1. Since, there is $\log(1 - c)$ term in the model equation, 0.999 is used as the maximum to avoid numerical failures. Volume averaging of Eq. 2 will still predict $\frac{dc_{avg}}{dt} = -\delta$. For 1 C charge (assuming $c = 0.001$ at the full charged state of the cell, corresponding to a low state-of-lithiation of the cathode), $\delta = \frac{1}{3600}$ gives the 1 C rate of charge. Model (2) has many attractive features compared to model (1), such as:

1. Model 2 does not need an extensive empirical fitting for open-circuit voltage expression (OCV) compared to model 1, which does need one. The presented model does need to be parameterized for ω and κ by fitting experimental data, but this is fundamentally and quantitatively different from the conventional approach to modeling OCV, which involves use of arbitrary functional relationships and fitting a much larger number of parameters. The voltage profile (assuming no kinetic limitation) is given by, $V = 3.42 - \frac{RT}{F}\mu_s$, where μ_s is the chemical potential at $x = 1$ in dimensionless form.
2. In certain materials, multiple phases need to be considered (due to phase separation or attraction) during intercalation. Tailoring the value of ω in model 2 is better suited and easier to implement to capture this phenomenon instead of an empirical profile obtained using experiments.¹³ Prof. Bazant has strongly advocated models of this type due to its thermodynamic consistency amongst other models developed in the last decade.
3. A simulation of model 2 at 25% cell state of charge is given in Figs. 1a and 1b for $\kappa = 0.001$ with $N = 100$ linear elements in COMSOL. For low and negative values of ω , model 2 predicts Fickian-like diffusion behavior as in model 1 but is much easier to simulate. For large values of ω ($\omega > 2$), proper care should be taken to simulate model 2, paying attention to numerical oscillations, convergence, and stability of the numerical scheme. These features make model 2 very rich in terms of physics and numerical analysis.

Limitations of the proposed model.—The model considered is one-dimensional. The effect of curvature will be seen only in 2D or 3D models. In addition, anisotropy is not included in the phase-field model reported in this paper. For model 2, explicit time stepping is difficult to achieve as the time step restriction will be based on $dt < h^4$ where h is the element/node size in spatial discretization. Implicit time stepping is feasible, but sometimes this might give meaningless results. When weak form FEM is used, linear, quadratic, and cubic are element shapes used in the simulation. They are referred to as first order (linear) and higher order (quadratic and cubic) shape functions in this paper. For example, the COMSOL simulation of this model for $\omega = -2$ and 4.5 for $N = 10$ and 100 elements with first order (linear) shape functions are shown in Figs. 1c–1f. One can see that concentration becomes negative and ideally the simulation should stop, but the stop condition might be skipped erroneously in default adaptive time-stepping methods with backward difference for time stepping (changing the solvers or tuning the parameters will avoid this problem). The oscillations at high values of ω do not go away with higher order methods or shape functions (quadratic or cubic) and they only fade away at a higher number of elements. Incidentally, as the number of elements is increased, convergence is seen, and only meaningful concentration values are predicted. Variable grid spacing with reduced mesh sizes near the surface, $x = 1$ might converge faster for this model. Model 2 features a log term for the description of μ variable. This makes the simulations susceptible to numerical inconsistencies. Therefore, a reformulation of the model equations is required as described in the next section. Finally, phase-field models are computationally challenging to solve and the theory to solve these models is still under development (in particular for robust and efficient simulation in CPU constrained environments).

Charging profiles and optimal control for reduced degradation.—Model-based BMS has shown significant improvement in battery life and reduced charging time for different battery chemistries and systems. There is a lot of uncertainty in battery degradation mechanisms and parameters. However, a meaningful objective for model 2 to minimize concentration gradients across a particle can be stated by using the formulation given below.

$$obj = \int_0^{t_f} (c_s - c(0, t))dt \quad [3]$$

Subjecting this to Eq. 2 we get $\int_0^{t_f} \delta = \frac{0.98}{3600}$. This objective minimizes the concentration gradient across the particle for a given total charge stored. A minimized concentration gradient within the particle will reduce stress induced degradation/cracking. The original model (Model 2) was reformulated as follows for ease of simulation and optimal control.

$$\begin{aligned} t_d \frac{\partial c}{\partial t} &= -\frac{\partial N_x}{\partial x}; N_x = -\left(c(1-c) \frac{\partial \mu^1}{\partial x} + \frac{\partial c}{\partial x}\right) \\ \mu^1 &= \omega(1-2c) - \kappa \frac{\partial^2 c}{\partial x^2} \\ x=0, \quad \frac{\partial c}{\partial x} &= 0; N_x = -c(1-c) \frac{\partial \mu^1}{\partial x} + \frac{\partial c}{\partial x} = 0 \\ x=0, \quad \frac{\partial c}{\partial x} &= 0; N_x = -c(1-c) \frac{\partial \mu^1}{\partial x} + \frac{\partial c}{\partial x} = \delta \cdot t_d \end{aligned} \quad [4]$$

To confirm that the reformulated model (Eq. 4) matches with the original model (Eq. 2), simulations were performed with increasing number of N , the number of elements. After $N = 100$, for all the values of ω , results are converged. Equation 4 does not have a logarithm term which reduces solver failures while running the model inside an optimization framework.

For obtaining the optimal control profiles a control vector parametrization approach was used with cell-centered finite difference formulation with a numerical method of lines (MOL) framework for Eq. 4. The number of stages (Nstages) is the number of intervals that the total charging time is divided into, in the optimal charging problem. The objective was found to monotonically decrease with an increase in Nstages when the number of stages was doubled as shown in Fig. 2a. The control profiles obtained for minimized degradation are given in Fig. 2b where $\omega = 4.5$.

Analysis of optimal control profiles.—The results from the control profiles suggest the following:

1. The improved model (Eq. 2) compared to diffusion model (Eq. 1) gives an impulse profile as the optimal control profile. This profile is also consistent with a recent experimental observation reported elsewhere.³⁹
2. Regular charging profiles at constant current charging (CC) for one hour show significant concentration gradients across the electrode due to phase separation (Fig. 2c). As opposed to CC charging, the optimal control profile significantly reduces the gradients across the electrodes as shown in Fig. 2c for $\omega = 4.5$.
3. The addition of more mechanisms (e.g., kinetics, mesoscale, or other detailed models) to model 2 might move the control profiles towards a pulse profile, as Professor Landau predicted earlier, or provide completely different control profiles.
4. An open-source framework is developed and shared for predicting the (im)pulse protocol reported in this paper.⁴⁰ This was first developed in MATLAB and future publications will explain the detailed math involved in optimizing the algorithms, for real-time simulation in embedded systems and BMS. In addition, a webpage will be maintained with simulation examples in Maple, MATLAB, and C.
5. The code presented can be modified to include kinetics and spherical coordinates. Nonlinear path constraints on voltage and temperature can be added as inequality constraints, but this

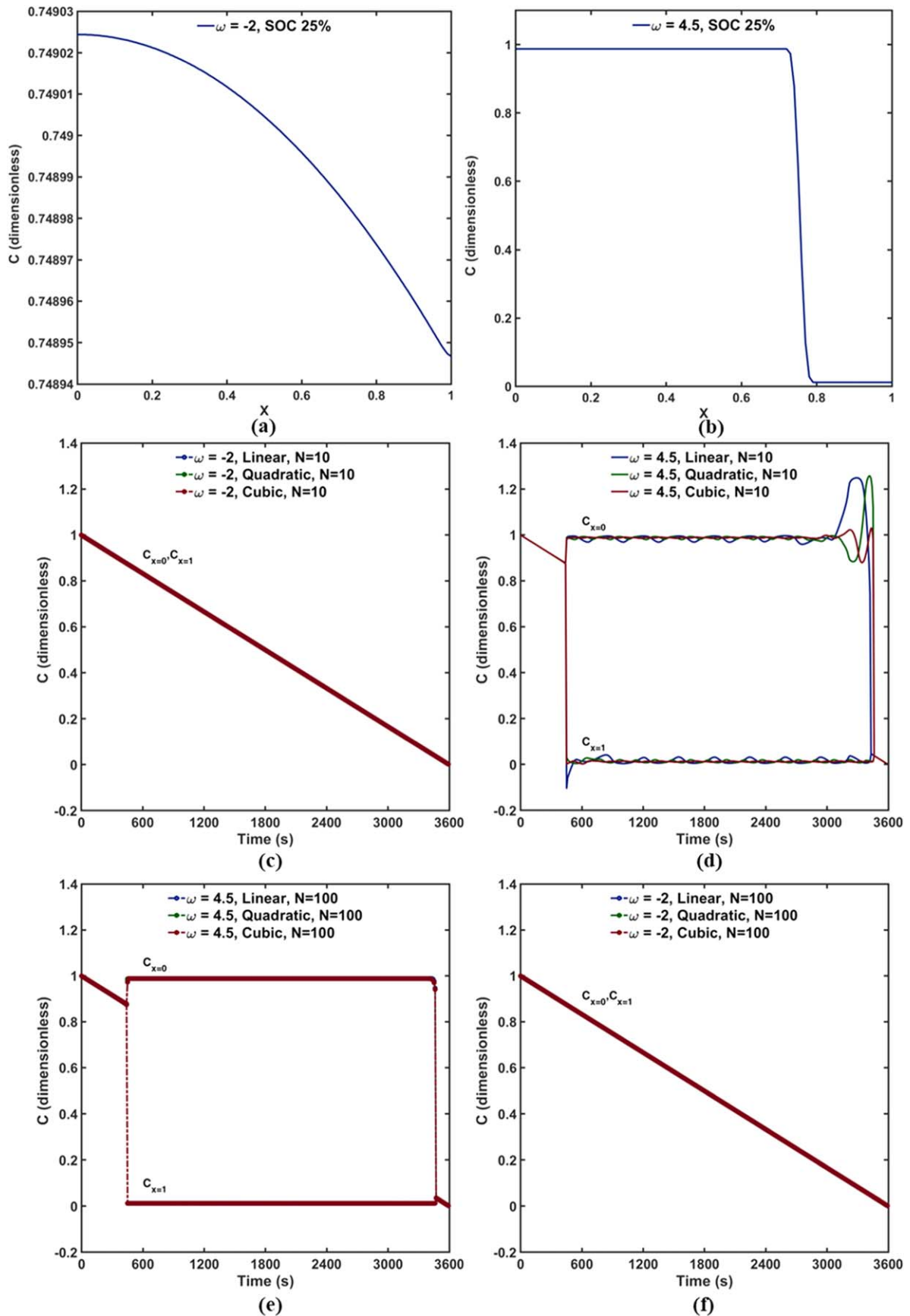


Figure 1. Results for model 2 for C_{surface} vs distance x for (a) $\omega = -2$, SOC = 25% (b) $\omega = 4.5$, SOC = 25%. Results for model 2 C_{surface} vs t for (c) $\omega = -2$ at $N = 10$ (d) $\omega = 4.5$ at $N = 10$ (e) $\omega = 4.5$ at $N = 100$, and (f) $\omega = -2$ at $N = 100$.

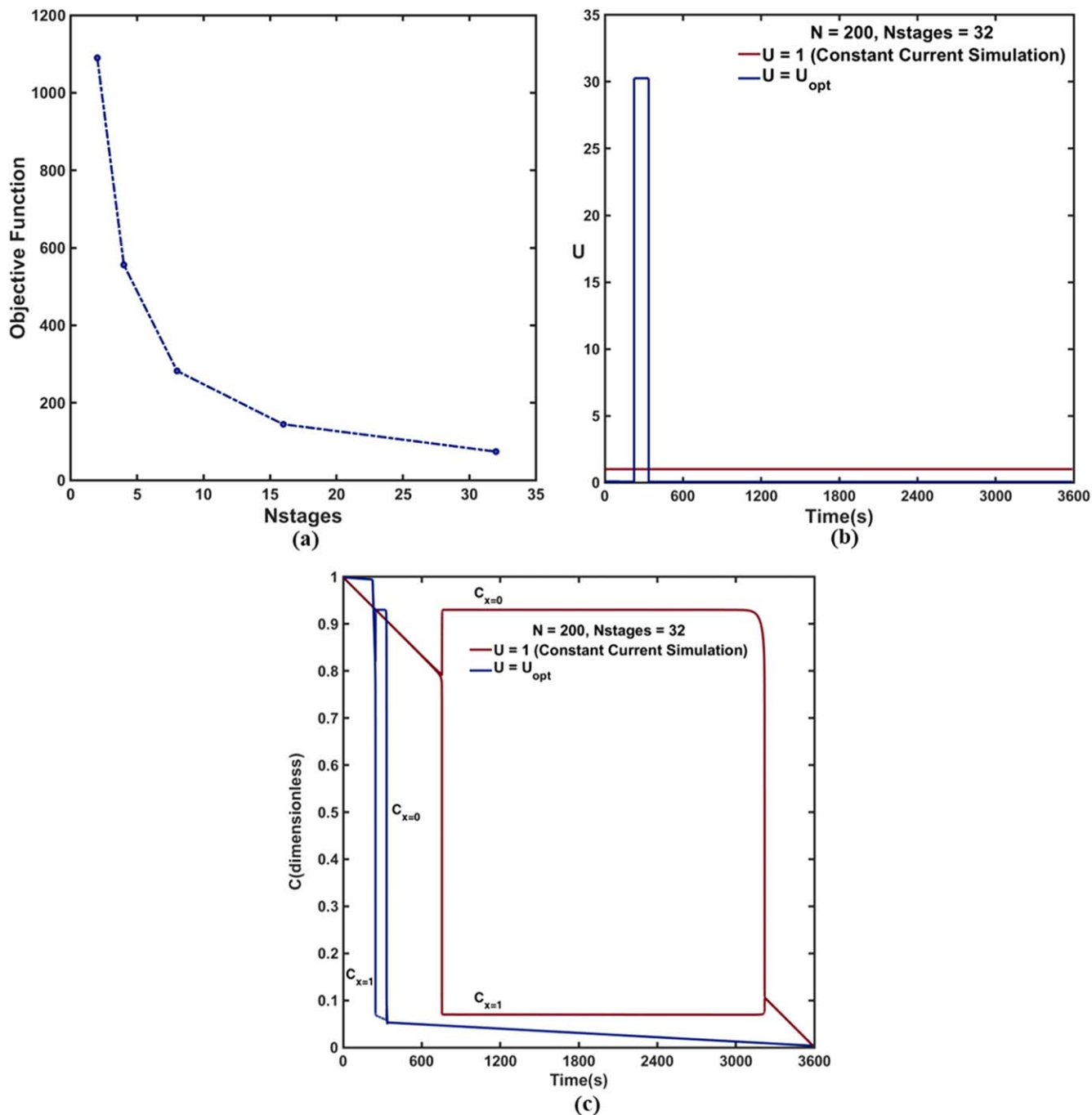


Figure 2. The control profiles obtained for minimized degradation for the phase-field model. (a) Objective function vs Nstages (number of stages)(Nstages is the number of intervals in which charging is divided to get optimal profile) (b) Constant current profile vs Phase-field model-based optimal charging profile for $\omega = 4.5$ (c) Concentration at $x = 0$ and $x = 1$ (surface) vs time for constant current vs phase-field model-based optimal charging for $\omega = 4.5$. The optimal control profile obtained for the model mimics an impulse profile.

might require additional convergence studies with respect to the number of control stages and number of node points.

Conclusions and future prospects.—Physics-based battery models are based on physical and electrochemical principles that govern the internal processes of the cell. As more and more physics is unearthed from innovative new diagnostic tools and techniques, there is a need to revisit the existing models. The case study shown in this paper suggests that when models are upgraded (in this paper, the diffusion equation was replaced with phase-field models), the optimal control profiles yield impulse-like behavior, which was

perhaps intuitively predicted by Professor Landau ahead of time.³¹ Conclusions and future prospects are summarized below:

1. *Physics:* Ideally, a thermodynamically consistent model should be chosen as opposed to models requiring empirical fits for the OCV expression (model 2 vs model 1). However, this requires additional computational work, and with the advent of GPU-based computers and improved software and hardware, more detailed multiscale models can be simulated efficiently and moved to control and design applications.
2. *Numerical simulation:* Grid convergence studies should be a part of numerical analysis when the physics is modified (for









example, model 1 is easier to simulate compared to model 2). Model 1 is amenable to spectral methods, but they might result in meaningless solutions for model 2. A simple cell-centered finite difference is used in the code for simulation and control. This was guided by the fact that higher order methods/shape functions did not remove the oscillations. The best possible numerical method depends on the physics and the operating conditions for the model. Rectangular coordinates were used in the paper and codes, and the numerical scheme used provides the same order of accuracy in spherical coordinates as well. If finite element methods and higher order methods are used, the spherical coordinates would require modifications at $x = 0$ for better accuracy and convergence. Additionally, the simulation approach was not optimized, however providing the structure and values of analytical Jacobian can speed up the simulation. Future work will involve efficient mathematical techniques to attain sufficient convergence with reduced CPU time possibly facilitated by reduced number of discretized equations (in space), different temporal discretization methods, parallel CPU/GPU computing, and other order reduction techniques.

- Software:** The results presented in this paper are based on MATLAB codes for simulation and optimization. Running the codes in C can result in a higher efficiency of simulation with lower memory footprint. Optimization of codes and algorithms for the model presented in this paper will be reported in future publications.
- Degradation mechanisms:** The results presented assume that higher currents do not lead to additional modes of degradation (thermal, SEI layer, Li plating, etc). Addition of other degradation or performance mechanisms can change the control profile obtained. This requires integration and validation of the control profiles with in situ diagnostic techniques.

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