

Accounting for the Effects of Probabilistic Uncertainty During Fast Charging of Lithium-ion Batteries

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Abstract—Batteries are nonlinear dynamical systems that can be modeled by Porous Electrode Theory models. The aim of optimal fast charging is to reduce the charging time while keeping battery degradation low. Most past studies assume that model parameters and ambient temperature are a fixed known value and that all PET model parameters are perfectly known. In real battery operation, however, the ambient temperature and the model parameters are uncertain. To ensure that operational constraints are satisfied at all times in the context of model-based optimal control, uncertainty quantification is required. Here, we analyze optimal fast charging for modest uncertainty in the ambient temperature and 23 model parameters. Uncertainty quantification of the battery model is carried out using non-intrusive polynomial chaos expansion and the results are verified with Monte Carlo simulations. The method is investigated for a constant current–constant voltage charging strategy for a battery for which the strategy is known to be standard for fast charging subject to operating below maximum current and charging constraints. Our results demonstrate that uncertainty in ambient temperature results in violations of constraints on the voltage and temperature. Our results identify a subset of key parameters that contribute to fast charging among the overall uncertain parameters. Additionally, it is shown that the constraints represented by voltage, temperature, and lithium-plating overpotential are violated due to uncertainties in the ambient temperature and parameters. The C-rate and charge constraints are then adjusted so that the probability of violating the degradation acceleration condition is below a pre-specified value. This approach demonstrates a computationally efficient approach for determining fast-charging protocols that take probabilistic uncertainties into account.

I. INTRODUCTION

Intercalation-based electrochemical batteries such as lithium-ion batteries (LIB)s are critical components for sustainable energy grids, transportation, and mobile devices. Among various battery materials, LIBs are the most widely used due to their characteristics of high energy density, high open-circuit voltage, low self-discharge characteristics, and long lifetime. Long charging times and reduced capacity due to battery deterioration, however, still remain challenges. As such, a goal of the battery industry is to develop technologies that enable fast charging while minimizing degradation.

Many battery models have been used to model the cycling behavior of LIBs. Each battery model can be categorized as being an equivalent circuit model (ECM) or an electrochemical model (EM). EMs describe internal phenomena such as temperature distribution, concentration distribution, and solid-electrolyte interface (SEI) growth during charging,

which is challenging for an ECM. The most widely used EM is the porous electrode theory (PET) model, also called the pseudo-two-dimensional (P2D) model [1]. Much research has been devoted to improving the modeling of degradation (e.g., [2], [3]). Further research has considered the efficient computational implementation of the PET model (e.g., [4] and citations therein) and on the lack of identifiability of some of the model parameters (e.g., [5], [6], and citations therein). The nominal PET model describes internal battery phenomena, without considering the uncertainties inherent in the cell-to-cell variation of lithium-ion batteries or variations in the battery environment. Uncertainty propagation should be considered when developing fast-charging protocols, however, to ensure that battery degradation remains limited when battery operation deviates from the nominal battery model.

Most studies on PET models ignore the effects of uncertainties on their predictions. A potential partial reason for this situation is because uncertainty quantification (UQ) that takes nonlinear dynamics into account via the Monte Carlo (MC) method requires on the order of ten thousand simulations for a single case study, so the computational time needed for the uncertainty analysis can be a bottleneck. The alternative approach of polynomial chaos theory evaluates uncertainty using an orthogonal polynomial expansion of the probability distribution of each random variable. Due to its significantly faster convergence speed and lower computational cost compared to the MC method, its application to battery applications has recently become of interest including applications for the estimation of state-of-health (SOH) and state-of-charge (SOC) [7], parameter sensitivity analysis [8], [9], [10], [11], parameter estimation [12], [13], and model predictive control [14].

Here, we consider the effects of uncertainty for the fast charging of batteries, and how this information can be used to design protocols that explicitly take probabilistic uncertainties into account. We use non-intrusive polynomial chaos expansion (PCE) to quantify the effects of uncertainty, identify confidential intervals (CIs) for the key states during charging, and compare the results to the nominal case to determine the impact of uncertainty.

This article is organized as follows. The next sections summarize the PET model and polynomial chaos theory. Next, we present a case study of the application of polynomial chaos theory to the PET model for the constant current–constant voltage (CC-CV) protocol, which is known to be nearly optimal for fast charging for this lithium-ion battery for operations restricted to satisfy the pre-specified

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current and voltage constraints [15]. We explicitly consider the effects of changing the time of the switching between the constant current (CC) phase and the constant voltage (CV) phase in response to the change in the ambient temperature and PET model parameters. That is, we take into account C-rate and charging constraints such that the probability of violating the degradation constraints is below a pre-specified value. The article concludes with a discussion of limitations and conclusions.

II. PET MODELING

The PET modeling framework for LIBs is a physics-based framework for modeling the cycling behavior of a cell. For LIBs, the model considers three regions: the porous anode, the electrolyte, and the porous cathode (Fig. 1). Diffusive transport is modeled by Fick's law and electrical resistance modeled by Ohm's law. The reaction at the interface between the liquid electrolyte and the solid particles is modeled by electrochemical interfacial reaction kinetics, usually by the Butler–Volmer equation. The resulting PET model is described by coupled partial differential-algebraic equations (DAEs). The PET model is also referred to as the P2D model, where the x dimension is the distance across the length of the cell and the r dimension is the so-called pseudo second dimension which describes diffusion inside porous electrode particles. More details of the model can be found in the original publication by Newman and coworkers [1].

When a load is connected to a charged LIB, some electrons leave the anode, powering the load, and some electrons enter the cathode. Inside the cell at the interface of porous graphite particle (anode, i.e., negative electrode), lithium ions enter the liquid electrolyte, leaving electrons behind, travel through the electrolyte and separator to reach a porous cathode particle (positive electrode), and electrochemically react again forming a lithium-metal-oxide, e.g., LiCoO_2 (Fig. 1). The complexity of the underlying reactions and potentially also parasitic reactions when degradation is considered leads to numerous model parameters, such as diffusion constants, reaction constants, and geometric electrode properties. A detailed nonlinear identifiability analysis indicates that many of the model parameters are uncertain when fit to the experimental data collected for lithium-ion batteries [5].

III. POLYNOMIAL CHAOS THEORY

UQ analyzes the impact of probabilistically uncertain parameters or inputs on the model states or outputs. The MC method is most commonly used in UQ. This method is based on statistics; when the number of samples is n , the convergence rate scales as $1/\sqrt{n}$ [16]. Accurately quantifying system uncertainty using the MC method requires many model evaluations, usually on the order of ten thousand or more, and is a computationally expensive task. Polynomial chaos theory is an alternative method to quantify the statistical information of the response while being orders of magnitude faster. As such, polynomial chaos theory is an interesting alternative to deal with the uncertainties of a complex dynamical system compared to the MC method. Polynomial chaos theory can

TABLE I
CORRESPONDENCE BETWEEN THE TYPE OF DISTRIBUTION AND
POLYNOMIAL BASIS

Distribution	Density function	Orthogonal polynomial	Hilbertian polynomial
Normal	$\frac{1}{\sqrt{2\pi}}e^{-x^2/2}$	Hermite $H_n(x)$	$H_n(x)/\sqrt{k!}$
Uniform	1/2	Legendre $P_n(x)$	$P_n(x)/\sqrt{12k+1}$

quantify the probability distribution function for any state, which can be plotted in the form of a histogram or can be used to construct prediction intervals for a variety of confidence levels for simpler visualization. Compared to the MC method, polynomial chaos theory has been shown to achieve similar accuracy in the quantification of the effects of probabilistic uncertainties while requiring much less computational cost [17].

Polynomial chaos theory uses orthogonal polynomials to quantify the effects of uncertainty on the system response caused by the effects of probabilistic inputs or parameters. Polynomial chaos theory can directly handle a large variety of distributions on the input parameters, including uniform and Gaussian, based on the Askey scheme [18]. Each distribution has a corresponding the optimal set of orthogonal polynomials as a basis, as shown in Table I.

Assume that the computationally intensive model M has n input parameters. The input vector is defined as $\mathbf{X} = \{x_{(1)}, x_{(2)}, x_{(3)}, \dots, x_{(n)}\}$ whose elements are assumed to be independent variables.

The univariate orthogonal polynomial determined according to the distribution of the input is expressed as a Hilbertian basis $\psi_k(x)$ through normalization [19], [20],

$$\Psi_\alpha(\mathbf{X}) = \prod_{i=1}^n \psi_{\alpha_i, (i)}(x_i). \quad (1)$$

Multivariate orthonormal polynomials, $\Psi_\alpha(\mathbf{X})$, can be obtained from the tensor products of the above univariate orthonormal polynomials $\psi_k(x)$,

$$M(\mathbf{X}) = Y = \sum_{\alpha \in \mathbb{N}^n} \mathbf{a}_\alpha \Psi_\alpha(\mathbf{X}). \quad (2)$$

The PCE for Y is defined for the input random vector \mathbf{X} and expands to the infinite series (2). In engineering applications, the series is reduced to being finite through truncation. The model reduced through the truncation follows the set $A^{n,p} = \{\alpha | \alpha \in \mathbb{N}^n, |\alpha| \leq p\}$, with $|\alpha|$ always less than or equal to the maximum degree p ,

$$\text{card } A^{n,p} \equiv P = \binom{n+p}{p} = \frac{(n+p)!}{n!p!}. \quad (3)$$

The reduced polynomial expansion Y^{PC} is represented by

$$M^{PC}(\mathbf{X}) = Y^{PC} = \sum_{\alpha \in A^{n,p}} \mathbf{a}_\alpha \Psi_\alpha(\mathbf{X}). \quad (4)$$

The coefficient of truncated PCE is calculated using a non-intrusive approach, where the least-squares minimization method was used [21]. This method identifies the coefficients

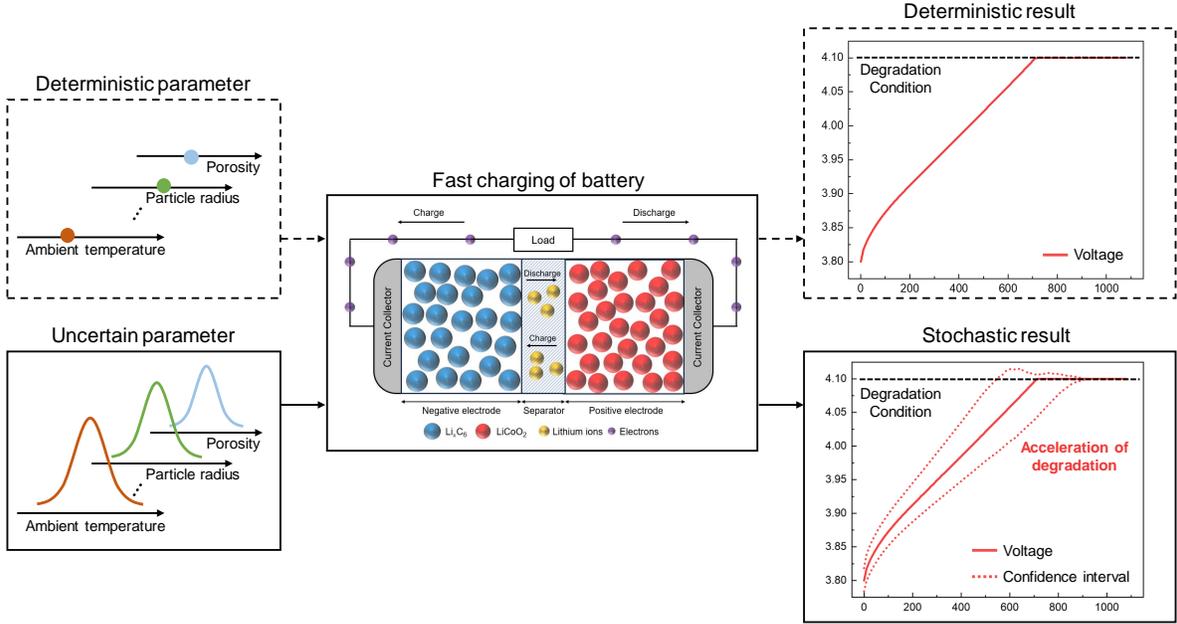


Fig. 1. Schematic of the uncertainty propagation during fast charging.

that minimize the truncation error of infinite and truncated polynomial expansions,

$$\hat{\mathbf{a}}_\alpha = \operatorname{argmin} \mathbb{E}[\mathcal{M}(\mathbf{X}) - \mathcal{M}^{PC}(\mathbf{X})]. \quad (5)$$

The uncertainty of the system is quantified using statistical moments calculated through polynomial coefficients, such as mean and variance, and each coefficient can be calculated through the orthonormality of the polynomial basis. The mean is expressed as the constant term a_0 of the polynomial expansion, and the variance is expressed as the sum of the squares of all coefficients excluding the constant term $\sum_{\alpha \in A^{n,p}} a_\alpha^2 - a_0^2$.

Moreover, the calculation of variance through simple computation of coefficients allows for variance-based global sensitivity analysis. The Sobol' indices is suitable for analyzing the sensitivity of parameters in complex systems such as LIB because it does not assume that the system is linear [22], [23]. Here, the analysis is done using total indices, which also take into account interactions between parameters.

IV. CASE STUDIES

This section considers the impact of probabilistic uncertainty propagating to the state of charge. The process of propagating from the input parameters of the PET model begins with computing the nominal simulation. There is evidence that high temperatures, voltages, and C-rates during battery charging accelerate battery degradation. Therefore, it is reasonable to introduce thresholds to minimize degradation: temperature (e.g., $> 40^\circ\text{C}$) and voltage (e.g., $> 4.1 \text{ V}$) [24], [25]. Additionally, side reactions cause lithium plating on the electrode, irreversibly reducing battery capacity, which is considered above 0 V because the lithium-plating overpotential accelerates as it decreases below 0 V.

We demonstrate the capabilities of PCE for quantification of the effects on uncertainties on the states for a CC-CV fast-charging protocol, which is known to be nearly optimal for the lithium-ion battery model considered in this study when subject to operating below maximum current and voltage constraints [15]. We also consider adjusting the transition time from constant current to constant voltage charging to ensure that the probability of violating the degradation constraints is below some pre-specified value.

A. CC-CV Charging

The effect of uncertain parameters during charging is identified through the CC-CV protocol. We analyzed charging states from 20% to 80% [26], [27], which are frequently considered the SOC range for fast charging of electronic vehicles. In CC-CV charging protocol, when the maximum voltage is reached in the CC phase, it switches to the CV phase and charging ends when SOC reaches 80% or the maximum temperature and minimum lithium-plating overpotential are reached.

Statistical information about the quantity of interest (QoI) of the three degradation conditions during CC-CV charging is obtained through PCE. If the R-squared (R^2) of the PCE is lower than 0.8, it is determined to have low reliability and is excluded from the results. 95% confidence interval (CI) of QoI analyzed at 10-second intervals for CC-CV charging are applied for three degradation conditions. Based on the charging protocol considering the CI, the constraints of the charging protocol and C-rate settings are discussed.

Figure 2 shows the total Sobol' indices for each degradation condition during CC-CV charging. For voltage, voltage control in CV mode reduces R^2 below 0.8. In that range, Sobol' indices and CI are not considered due to the low reliability of PCE. Among the 24 uncertain parameters specified

TABLE II
DESCRIPTION OF UNCERTAIN PARAMETERS [11]

Parameter	Description	Unit	Reference value	Random input
T_{amb}	Ambient temperature	K	298.15	Gaussian, $\mu = 298.15$, $\sigma = 1.0$
D_p^s	Positive solid-phase diffusivity	m^2s^{-1}	1.0×10^{-14}	Uniform, $[0.9 \times 10^{-14}, 1.1 \times 10^{-14}]$
D_n^s	Negative solid-phase diffusivity	m^2s^{-1}	3.9×10^{-14}	Uniform, $[3.51 \times 10^{-14}, 4.29 \times 10^{-14}]$
k_p	Positive reaction rate constant	$m^{2.5}mol^{-0.5}s^{-1}$	2.334×10^{-11}	Uniform, $[2.1 \times 10^{-11}, 2.56 \times 10^{-11}]$
k_n	Negative reaction rate constant	$m^{2.5}mol^{-0.5}s^{-1}$	5.031×10^{-11}	Uniform, $[4.52 \times 10^{-11}, 5.53 \times 10^{-11}]$
D_p	Positive electrolyte diffusivity	m^2s^{-1}	7.5×10^{-10}	Uniform, $[6.75 \times 10^{-10}, 8.25 \times 10^{-10}]$
D_s	Separator electrolyte diffusivity	m^2s^{-1}	7.5×10^{-10}	Uniform, $[6.75 \times 10^{-10}, 8.25 \times 10^{-10}]$
D_n	Negative electrolyte diffusivity	m^2s^{-1}	7.5×10^{-10}	Uniform, $[6.75 \times 10^{-10}, 8.25 \times 10^{-10}]$
L_a	Positive current collector thickness	m	1.0×10^{-5}	Uniform, $[0.8 \times 10^{-5}, 1.2 \times 10^{-5}]$
L_p	Positive electrode thickness	m	8.0×10^{-5}	Uniform, $[7.7 \times 10^{-5}, 8.3 \times 10^{-5}]$
L_s	Separator collector thickness	m	2.5×10^{-5}	Uniform, $[2.2 \times 10^{-5}, 2.8 \times 10^{-5}]$
L_n	Negative electrode thickness	m	8.8×10^{-5}	Uniform, $[8.5 \times 10^{-5}, 9.1 \times 10^{-5}]$
L_z	Negative current collector thickness	m	1.0×10^{-5}	Uniform, $[0.8 \times 10^{-5}, 1.2 \times 10^{-5}]$
ϵ_p	Positive porosity	-	0.385	Uniform, [0.36, 0.41]
ϵ_s	Separator porosity	-	0.724	Uniform, [0.63, 0.81]
ϵ_n	Negative porosity	-	0.485	Uniform, [0.46, 0.51]
R_p^p	Positive particle radius	m	2.0×10^{-6}	Gaussian, $\mu = 2.0 \times 10^{-6}$, $\sigma = 0.3896 \times 10^{-6}$
R_n^p	Negative particle radius	m	2.0×10^{-6}	Gaussian, $\mu = 2.0 \times 10^{-6}$, $\sigma = 0.1354 \times 10^{-6}$
Brugg _p	Positive Bruggeman coefficient	-	4.0	Uniform, [3.8, 4.2]
Brugg _s	Separator Bruggeman coefficient	-	4.0	Uniform, [3.8, 4.2]
Brugg _n	Negative Bruggeman coefficient	-	4.0	Uniform, [3.8, 4.2]
t_+	Transference number	-	0.364	Uniform, [0.345, 0.381]
σ_p	Positive electronic conductivity	$S m^{-1}$	100	Uniform, [90, 110]
σ_n	Negative electronic conductivity	$S m^{-1}$	100	Uniform, [90, 110]

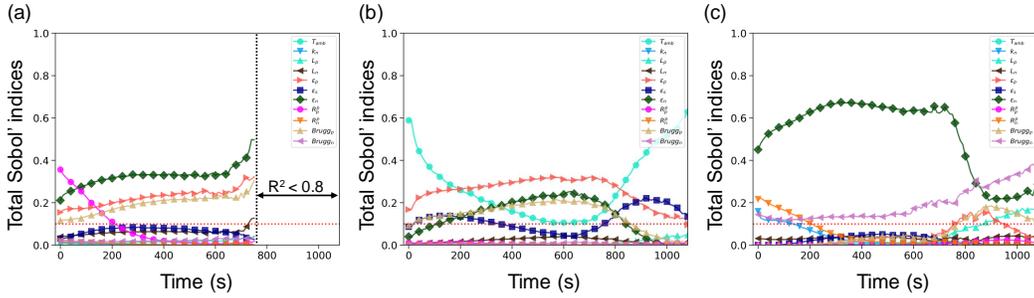


Fig. 2. Total Sobol' indices for three degradation conditions for 2.2C CC-CV charging: (a) voltage, (b) temperature, (c) lithium-plating overpotential.

in Table II, 11 high sensitivity parameters (Sobol' indices > 0.1) for the three degradation conditions are identified as

- Voltage: L_n , ϵ_p , ϵ_n , R_p^p , Brugg_p
- Temperature: T_{amb} , ϵ_p , ϵ_s , ϵ_n , Brugg_p
- Lithium-plating overpotential: k_n , L_p , ϵ_p , ϵ_n , R_n^p , Brugg_p, Brugg_n

The union of the sets determined with highly sensitive parameters for each condition was identified as the key parameter set for accelerated degradation.

Figure 3abc shows the nominal results and CI of voltage, temperature, and lithium-plating overpotential for 2.2C CC-CV charging of $LiC_6/LiCoO_2$ depicted through the parameters of Ref. [28]. Non-intrusive PCEs are generated using 300 samples. In addition, the QoI approximated by PCE for the three degradation conditions near the time of switching to the CV stage is evaluated at 10,000 samples and compared to MC using 3,000 samples (Fig. 3def). According to Table III, PCE generated through key parameters takes only about 10.1% of the computational budget compared to MC.

The nominal model indicates that the charging should be switched to CV mode in about 711 seconds when V reaches

TABLE III
COMPUTATIONAL TIMES IN CC-CV APPLICATIONS BETWEEN MONTE CARLO, PCE USING 24 PARAMETERS, AND PCE USING 11 PARAMETERS AT 600 SECONDS

	MC (24 parameters)	PCE (11 parameters)
Time (s)	3,997	403

V_{max} (Fig. 3a). The upper bound of the voltage CI reaches V_{max} at about 550 seconds. As such, in battery modules or packs composed of many cells, voltage constraints may be reached locally due to cell variability, which may lead to uneven degradation. Skewness is observed in the distribution as the CV mode is approached, which results in the CI by PCE overestimating the V_{max} (Fig. 3d). The nominal temperature rises to 311.4 K during charging, which does not reach the accelerated degradation condition, but the CI exceeds the T_{max} of 313.15 K from about 530 to 930 seconds (Fig. 3b). The voltage switches to the CV phase when it reaches V_{max} , but charging is terminated when the temperature reaches T_{max} to minimize performance degradation such as irreversible decomposition of the electrolyte. For

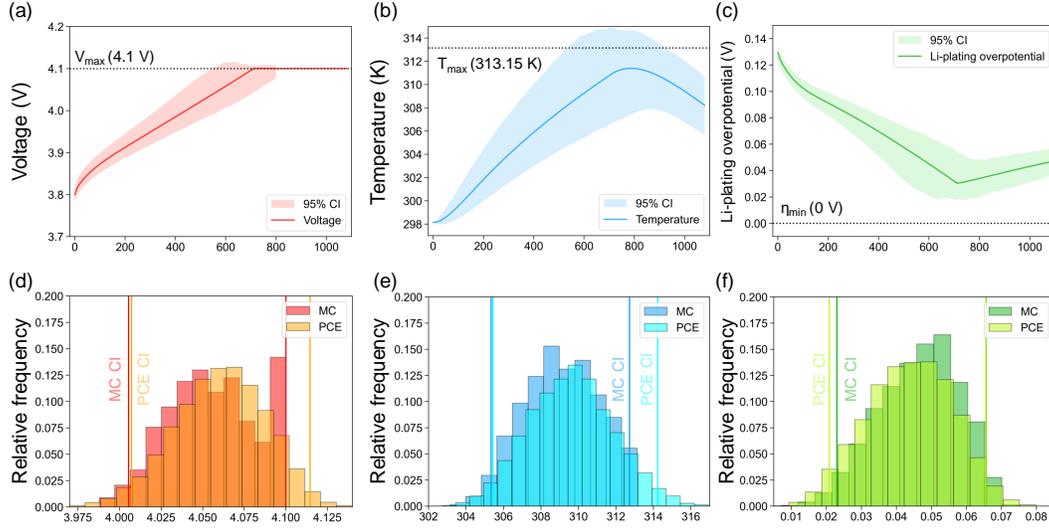


Fig. 3. Nominal values and CI for three degradation conditions in 2.2C CC-CV: (a) voltage, (b) temperature, (c) lithium-plating overpotential. QoI comparison by MC and PCE at 600 seconds: (d) voltage, (e) temperature, (f) lithium-plating overpotential.

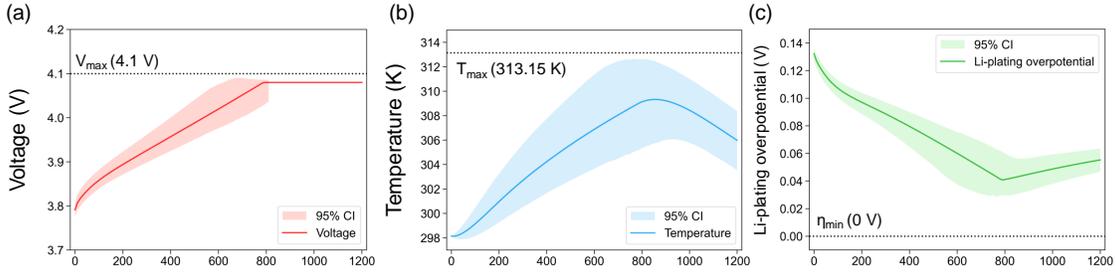


Fig. 4. Nominal values and CI for three degradation conditions in 2.0C CC-CV ($V_{\max} = 4.08$ V): (a) voltage, (b) temperature, (c) Li-plating overpotential.

this reason, a truncated distribution rather than skewness is observed for temperature, and the lowered R^2 is the cause of the upper bound of CI predicting a value of temperature higher than T_{\max} (Fig. 3e). Degradation does not occur due to the T_{\max} constraint, but the desired SOC (i.e., 80%) is not reached as charging termination. In other words, uncertainty propagation in 2.2C CC-CV charging indicates that degradation is accelerated by voltage, or charging is prematurely terminated by reaching temperature constraints. Since the lithium-plating overpotential does not reach the constraint during charging, the distributions identified by MC and PCE during charging are quite similar (Fig. 3f).

Figure 4abc shows that the degradation condition is not reached for C-rate of 2C. As V_{\max} decreases to 4.09 V, the upper bound of the CI does not reach 4.1 V. Reducing the C-rate to 2C also reduces the temperature rise so that the temperature CI does not reach T_{\max} . Unlike charging at 2.2C, the upper bound of temperature CI for a C-rate of 2C does not show a significant difference compared to CI by MC (Fig. 5). The C-rate-dependent lithium-plating overpotential

TABLE IV
COMPARISON OF CHARGING TIME AND ACCELERATED DEGRADATION ACCORDING TO C-RATE AND V_{\max} FOR CC-CV CHARGING

C-rate	2.2C	2.0C
V_{\max} (V)	4.1	4.08
CC to CV (s)	711.2	787.3
Total charging time (s)	1086.2	1204.8
Degradation	accelerated	accelerated

likewise does not reach the constraint (Fig. 4c). However, charging under moderate conditions increases the charging time by about 118 seconds (Table IV).

V. CONCLUSION AND FUTURE DIRECTION

This study analyzes the impact of uncertainty on battery degradation during charging using the PET model which expresses various internal phenomena of a battery. The PET model has been shown to be effective in modeling LIBs, but its nominal predictions can be sensitive uncertainties in environmental and model parameters. PCE is applied to CC-CV charging to identify parameters sensitive to fast

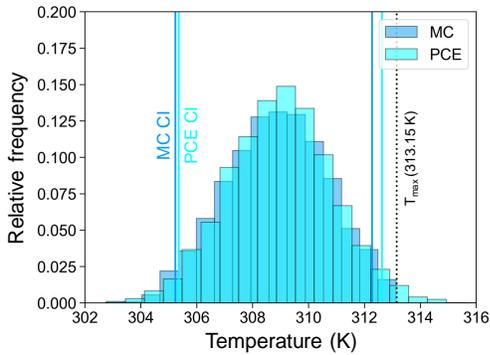


Fig. 5. Probability distribution and CI by MC and PCE at 800 seconds in 2.0C CC-CV charging.

charging conditions and investigate their effect. Among the 24 parameters consisting of ambient temperature and 23 PET model parameters, only 11 parameters were identified to affect the charging status. In nominal results, once the maximum voltage is reached in the CC stage, the degradation is not accelerated due to the transition to the CV stage. On the other hand, when temperature and lithium-plating overpotential reach constraints during charging, charging is terminated. Uncertainties propagated to the degradation conditions indicate that accelerated degradation and premature charge termination may occur, which were not observed in the nominal results. Stochastically accelerated degradation can be minimized through adjustment of charging constraints such as V_{\max} or C-rate. Our results indicate that uncertainty during charging should be considered to minimize battery performance degradation. This non-intrusive PCE-based approach can extract statistical information of QoI with a significantly lower computational budget than MC and can be successfully applied to further improved and complex state-dependent charging protocols.

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