

Reduced-Order Model of Lithium-Iron Phosphate Battery Dynamics: A POD-Galerkin Approach

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Abstract—Lithium iron phosphate batteries with plateau in the open circuit voltage, hysteresis, and path dependence dynamics due to phase transition during intercalation/de-intercalation are challenging to model and even more challenging to control. A core-shell electrochemical modeling approach is able to capture the phase transition behavior at the cost of using a fine-grained spatial grid to transform the governing Partial Differential Algebraic Equations into Ordinary Differential Algebraic Equations, resulting in a computationally expensive system intractable for the design of real-time battery management system algorithms. This letter presents a reduced-order modeling paradigm to transform the high-dimensional model into a low-dimensional yet accurate control oriented electrochemical model. The Proper Orthogonal Decomposition-Galerkin method is used to reduce the state variable vector from 169 to a meager 9 with negligible loss in fidelity. The reduced-order model is validated against both experimental data and the highdimensional model for discharging-charging load profiles of different C-rates and real driving cycles. Promising results with one-third the computational burden and a voltage RMS error of less than 0.6% are achieved.

Index Terms—Model/controller reduction, energy systems, modeling.

I. INTRODUCTION

WHILE high energy density lithium-ion battery (LIB) technology continues to become the mainstay of electrified mobility and renewable energy solutions, we may face some grave challenges in the future in the form of supply-chain issues of cobalt and increased demand on nickel [1].

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Cobalt- and nickel-free positive electrode materials for LIBs in the form of Lithium Iron Phosphate (LFP) are currently being considered for mass production in electric vehicle batteries. Despite their lower energy density, LFP batteries offer good electrochemical stability, thermal performance, and cycle life making them a suitable candidate for cobalt- and nickel-free batteries [2]. Olivine structured LFP batteries are characterized by a flat open circuit voltage due to the phase transition [3] and are accompanied by a significant hysteresis between the open circuit voltage during charge and discharge and strong path dependence dynamics [4], making it challenging to model and predict their internal behavior accurately. In particular, the dreaded flat voltage plateau is a major challenge to designing algorithms for real-time Battery Management System (BMS). Owing to that, an electrochemical model that can accurately capture the battery dynamics, especially the phase transition behavior, would prove to be a good basis for designing advanced BMS algorithms.

Among the electrochemical models that take into account the phase transition behavior of LFP batteries, the core-shell technique is chosen in this letter. First developed by [5], it has been used in [6] and [7] and most recently in [8], [9]. In [10], a core-shell Enhanced Single Particle Model (ESPM) is combined with a machine learning-based hysteresis model, to form a *hybrid model* to accurately capture the hysteresisinduced voltage behavior and the path dependence dynamics under real world drive cycles. Battery dynamics governed by Partial Differential Algebraic Equations (PDAEs) are spatially discretized into Ordinary Differential Algebraic Equations (ODAEs) via Finite Difference Method (FDM) and Finite Volume Method (FVM), resulting in computationally expensive models not conducive for real-time BMS algorithm design. For instance, the mass conserving core-shell ESPM in [8] was discretized into an ODAE model with 169 state variables and running that model on a hardware-constrained on-board microcontroller is a difficult proposition. Hence, there is a need for a low-dimensional model that can capture the dominant behavior of the system and be accurate yet computationally inexpensive.

Various reduced-order model (ROM) techniques have been proposed to reduce the model dimensionality of LIB electrochemical models [11], such as Padé approximation [12], [13], proper orthogonal decomposition (POD) [14], [15], and residue grouping [16]. However, the resulting ROMs do not account for phase transition dynamics. It is to be noted that a core-shell ESPM for LFP is governed by nonlinear coupled

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dynamics due to the inclusion of one Ordinary Differential Equation (ODE) in the positive electrode that models the moving boundary indicating the phase transition. Such an additional moving boundary ODE makes the model unsuitable for popular reduction methods such as Padé approximation or balanced model reduction/truncation. In [7], the solid state diffusion dynamics of a core-shell model is approximated by a polynomial to obtain a ROM. Another alternative approach is to utilize coarse-grained spatial grid points, instead of fine-grained, while discretizing battery governing PDAEs into ODAEs. However, fewer grid points result in a low-dimensional feeble system that is not only inaccurate but also susceptible to lack of numerical convergence while solving the moving boundary ODE that indicates the phase transition.

To that end, the *contributions* of this letter are: 1) a mathematical formulation to reduce the high-dimensional average core-shell ESPM (CSa-ESPM) from [9] into a low-dimensional model via the POD–Galerkin method; 2) validation of the resulting state space ROM against the high-dimensional model and experimental data under both constant current (CC) and real driving cycle conditions.

This letter is structured as follows: in Section II, the governing equations of the CSa-ESPM are introduced. Section III describes the POD-Galerkin method. Section IV shows the ROM formulation for the CSa-ESPM and simulation results validating the ROM against the full order model and experimental data from a 49 Ah LFP battery. Finally, conclusions are summarized in Section V.

II. AVERAGE CORE-SHELL ESPM

In this section, the physical working principles of the graphite/LFP battery and the CSa-ESPM electrochemical governing equations [9] are briefly reviewed. The key aspect of the CSa-ESPM electrochemical model is the description of the phase transitions in the positive electrode during charge and discharge while representing an advanced version of the traditional core-shell ESPM [8], in which the discontinuities in the positive electrode open circuit potentials (OCPs) and overpotential during the phase transition are removed.

LFP electrode experiences three main 'phases' during charging and discharging [3], namely: 1) a Li-rich phase, LiFePO₄, denoted as β -phase, 2) a two-phase transition where LiFePO₄ and FePO₄ coexist, and 3) a Li-poor phase, FePO₄, denoted as α -phase.

Fig. 1 provides a schematic representation of these three phases within the model for a constant discharge cycle from 100% to 0% of SOC. Both electrodes are modeled as spherical particles, where the negative electrode is always in single phase whereas the positive electrode can be in one-phase (1P) or two-phase (2P). Further, \bar{t} and \bar{t}_f highlighted in the Fig. 1, refer to the initial and final time of the 2P condition. When the battery is fully charged, the positive electrode is in the α -phase (Li-poor), and it stays in that phase (1P) from when the discharge starts (Li-ions starts intercalate into the positive electrode) until the positive electrode reaches the normalized concentration θ^{α} at time instant $t = \bar{t}$. At this point, the β phase starts forming on the outer shell of the positive particle and the two phases (α and β) coexist. The core-region in α -phase is subjected to a shrinking process, while the thickness of the shell-region, in β -phase, increases as the battery is being discharged. When the transition from the α -phase



Fig. 1. Constant current discharge voltage profile. The moving radius (r_p) is reported to illustrate the positive electrode one-phase ($r_p/R_{s,p} = 0$) and two-phase ($r_p/R_{s,p} > 0$) regions. The times \bar{t} and \bar{t}_f denote the initial and final time instant of the two-phase condition.

to β -phase is over at time $t = \bar{t}_f$, the entire positive particle remains in β -phase until the end of the discharge. The inverse process takes place during the charging cycle (from β phase to α -phase). The Li-ion concentration in the negative, as well as in the 1P and in the 2P positive electrode shellregion, is described by the Fick's diffusion law, as shown in [8, Table 1]. The core-region of the positive electrode remains at a constant and uniform concentration equal to $c_{s,p}^{\alpha}$ ($c_{s,p}^{\beta}$) during discharging (charging). Further, for $t \in [\bar{t}, \bar{t}_f]$ [8, eq. (73)] describes the dynamics of the moving boundary r_p , describing the distance between the center of the positive particle and the interface between the two phases. Note that the motion of r_p depends on the concentration gradient $(\partial c_{s,p}/\partial r)$ across the α -phase and β -phase interface. Finally, the mass and the charge conservation within the electrolyte, are modeled as in [8, Table 1].

A. CSa-ESPM: State-Space Formulation

The cell electrochemical dynamics modeled as a set of nonlinear coupled PDAEs are numerically solved using the FDM and FVM, for the solid and liquid phase, respectively [17]. The discretization meshes used to approximate the Li-ion diffusion dynamics within the electrodes are illustrated in Fig. 1. The radius of the negative electrode particle, and the radius of the positive electrode particle for the 1P and the shell-region, are divided into N_r discretization points. Meanwhile, each cell domain (i.e., n: negative electrode, s: separator and p: positive electrode) is discretized into N_x volumes for the electrolyte dynamics approximation, as shown in Fig. 1. Following [17], the mass conservation in the solid phase is approximated into a system of $(N_r - 1)$ ODEs, as shown in (1) and (3), for the negative electrode and 1P positive electrode, respectively. On the other hand, (4) and (5) approximate the diffusion dynamics in the 2P positive electrode shell region and the moving boundary motion, where g(I) is equal to $c_{s,p}^{\beta}$ and $c_{s,p}^{\alpha}$ for

the discharge and charge, respectively. It is worth noting that Ffin (4) a time-varying grid size is considered to account for the variation of the shell region thickness during cycling, as widely described in [8]. Finally, the mass conservation in the liquid phase is discretized into a system of $3N_x$ ODEs (2), as discussed in [18]. The detailed state-space model description, including the model matrices and coefficients, is provided in [8]. Note that the model state vectors are the Li-ion concentration in the electrodes $(c_{s,j} = [c_{s,j,1} \cdots c_{s,j,N_r-1}]^T \in \mathbb{R}^{N_r-1})$ and in the electrolyte $(c_e = [c_{e,n} c_{e,s} c_{e,p}]^T \in \mathbb{R}^{3N_x}$ where $c_{e,j} = [c_{e,j,1} \cdots c_{e,j,N_x}] \in \mathbb{R}^{N_x}$). The $c_{s,j,i}$ refers to the Liion concentration of the i-th discretization point within the j-th electrode (wherein $j \in [n,p]$ in solid phase), while $c_{e,j,i}$ indicates the Li-ion concentration of the i-th volume of the j-th cell domain (wherein $j \in [n,s,p]$ in electrolyte phase). Moreover, $c_{s,j}^{suf} = c_{s,j,N_r-1}$ and $c_{s,j}^{bulk} = \frac{3}{R_{s,j}^3} \int_0^{R_{s,j}} r^2 c_{s,j} dr$ refer to the electrode surface and bulk concentration, respectively. Finally, the overall cell voltage $(V_{cell}^{CC} \text{ and } V_{cell}^{DV})$ is obtained as in (6) for CC and real driving cycles, respectively. In par-ticular, V_{cell}^{DV} accounts for the extra term V_h which models the hysteresis and the path dependence dynamics when the battery undergoes a real driving cycle. In this letter, V_h is provided by a physics-informed feedforward neural network (FNN), that was trained and validated in [10]. The FNN is composed by 9 hidden layers with 17 neurons for each layer, and uses I_{cell} , and $\theta_p^{bulk} = c_{s,p}^{bulk}/c_{s,p}^{max}$ and c_e^{avg} from the solution of the CSa-ESPM, as input features [10].

 U_j and η_j are the electrode OCP and overpotential, respectively, I_{cell} is the input cell current, $\Delta \Phi_e$ is the electrolyte overpotential and R_l is the lumped cell resistance. U_n and η_n reported in (7) and (9) depend only on the particle surface concentration, while the positive electrode quantities (U_p and η_p) are calculated as a function of $c_{s,p}^{bulk}$ during the 2P condition and $c_{s,p}^{surf}$ during the 1P one, as shown in (8) and (9). According to [9], the $c_{s,p}^{bulk}$ is utilized during the 2P positive electrode to address the discontinuity in the output due to the surface concentration during the phase transition.

Finally, the average positive OCP $((U_p^{ch} + U_p^{dis})/2)$ is used in V_{cell}^{DV} as opposed to U_p used in V_{cell}^{CC} . The SOC of each electrode is calculated based on the normalized bulk concentration (θ_i^{bulk}) , as in (12).

(θ_j^{bulk}), as in (12). It is important to note that the transition from one to twophase region ($t = \bar{t}$) occurs when θ_p^{bulk} reaches $\theta_p^{\alpha} = c_{s,p}^{\alpha}/c_{s,p}^{max}$ ($\theta_p^{\beta} = c_{s,p}^{\beta}/c_{s,p}^{max}$) for discharging (charging). At the following time instant, the moving boundary switches from 0 to $R_{s,p} - \epsilon$ since the two-phase interface is on the particle surface, where ϵ is a small constant utilized to prevent singularities in the model formulation. The r_p then begins to decrease as a result of the core-region shrinking process.

III. POD-GALERKIN MODEL ORDER REDUCTION

The POD-Galerkin method is a model order reduction (MOR) technique commonly used to approximate highdimensional system of ODAEs arising from the semidiscretization of PDAEs. It allows the reduction of the state space vector of the spatially discretized model maintaining its essential features and accuracy. In this section, the theoretical formulation of the POD-Galerkin MOR is briefly described, according to [19]. Considering the dynamical system written as:

$$\begin{cases} \dot{x}(t) = Ax(t) + f(x(t)) \\ x(t_0) = x_0 \end{cases}$$
(13)

where $x \in \mathbb{R}^n$, $A \in \mathbb{R}^{n \times n}$, $f : \mathbb{R}^n \to \mathbb{R}^n$ is a vector field and $x_0 \in \mathbb{R}^n$ is the initial condition. Note that the considered system is composed by a linear part, Ax(t), and a non-linear part, f(x(t)). The solution $x_k \in \mathbb{R}^n$ of (13) for a given time instant $t_k \in [0, +\infty]$ is called **Snapshot**. Given a sequence of Snapshot x_k , obtained for a given time mesh $t_k = t_0 + k \cdot dt$ where dt is the sampling time and $k = 0, 1, \ldots, n_t$, the **Snapshot matrix** $(S \in \mathbb{R}^{n \times n_t})$ is constructed as $S = [x_0 \ x_1 \ \cdots \ x_{n_t}]$. The key aspect of the POD is that it allows to extract an orthonormal basis $[\psi_1, \ldots, \psi_z]$ of a subspace $\Psi \subset \mathbb{R}^n$ with dimension z < n such that the distance between the Snapshot matrix and its projection on the new basis functions is minimized:

$$\min_{\psi_1 \cdots \psi_r} \sum_{i=0}^{n_t} ||x_i - \sum_{j=1}^{z} \langle x_i, \psi_j \rangle \psi_j ||^2,$$
(14)

where $\langle \cdot, \cdot \rangle$ is an inner product on Ψ such that $\langle \cdot, \cdot \rangle = \|\cdot\|^2$, $\langle \psi_i, \psi_i \rangle = 0$ if $i \neq j$ and $\langle \psi_i, \psi_i \rangle = 1$ if i = j. The optimal basis which solves the optimization problem (14) can be obtained through the Singular Value Decomposition (SVD) of the Snapshot matrix. In particular, the SVD allows to rewrite the snapshot matrix as $S = V \Sigma W^T$ where $\Sigma \in \mathbb{R}^{n \times n_t}$, $V \in \mathbb{R}^{n \times n}$ and $W \in \mathbb{R}^{n_t \times n_t}$. The columns of V are called **POD modes** and provide the optimal basis function $|\psi_1, \ldots, \psi_n|$, while the matrix Σ contains a non-null diagonal sub-matrix of dimension $n \times n$ in which the singular values (σ_i) are stored in a decreasing manner, namely $\sigma_1 > \sigma_2 > \cdots > \sigma_n > 0$. Finally, the truncation of model dynamics is computed by keeping the first r dominant modes $V_r = [\psi_1, \dots, \psi_r]$ of the optimal basis function, to maintain the main features of the system. A common approach to select the truncation order (r) is by defining the **truncation degree** (Θ_r) , calculated as $\Theta_r = (\sum_{i=1}^r \sigma_i^2) / (\sum_{i=1}^n \sigma_i^2)$. Note that, Θ_r needs to be close to 1 to approximate the solution of the model accurately. Finally, the Galerkin projection of the system (13) onto the new subspace V_r is used to reduce the dimension of the state-space model. The ROM is written as:

$$\begin{cases} \tilde{x}(t) = V_r^T A V_r \tilde{x}(t) + f(V_r \tilde{x}(t)) \\ \tilde{x}(t_0) = V_r^T x_0, \end{cases}$$
(15)

where $\tilde{x} \in \mathbb{R}^r$ is the state vector of the ROM. It worth mentioning that an approximation $(\bar{x}(t) \in \mathbb{R}^n)$ of the original state vector x(t) is reconstructed as $x(t) \approx \bar{x}(t) = V_r^T \tilde{x}(t)$.

IV. POD-GALERKIN REDUCED ORDER MODEL

In this section, the POD-Galerkin based ROM is formulated for the discretized CSa-ESPM (Table I), according to Section III. Fig. 2 provides a schematic representation of the steps required to develop the ROM and Table II reports the mathematical formulation of the model. According to (17–20), four different Snapshot matrices are generated solving (1–4) for a given constant input current. The columns of S_n and S_e contain the solution vectors $c_{s,n}$ and c_e of (1) and (2), for each time instant from the beginning (t_0) to the end (t_{end}) of the charge/discharge. Meanwhile, $S_{p,1P}$ and $S_{p,2P}$ are constructed considering the positive electrode concentration ($c_{s,p}$) during 1P ($t \in [t_0, \bar{t} - 1] \vee [\bar{t}_f + 1, t_{end}]$) and 2P ($t \in [\bar{t}, \bar{t}_f]$), respectively. Then, the truncated POD basis functions ($V_n^r, V_{p,1P}^r, V_{p,2P}^r$ and V_e^r) are obtained by



TABLE I

Fig. 2. Model reduction flowchart applied to the CSa-ESPM

keeping the first dominant columns of the basis functions $(V_n, V_{p,1P}, V_{p,2P} \text{ and } V_e)$ that were previously generated by computing the SVD of the snapshot matrices, as reported in (21). Different truncation orders $[r_e, r_n, r_{p,1P}, r_{p,2P}]$ are selected for the electrolyte, negative and positive electrode in both 1P and 2P, respectively, such that the truncation degrees result close to 1, $r_e < N_x$ and $r_n, r_{p,1P}, r_{p,2P} < N_r$. Finally, the original state-space model (1–4) is projected onto new subspaces as shown in (22) in accordance with (15). The new state vectors, indicated with $\tilde{c}_{s,n} \in \mathbb{R}^{r_n}, \tilde{c}_{s,p} \in \mathbb{R}^{r_{p,P-2P}}$ and $\tilde{c}_e \in \mathbb{R}^{r_e}$, correspond to the projection of the Li-ion concentration in both electrodes and in the electrolyte onto the new subspaces. Note that the reduced-order state-space model is

composed by $(r_n + r_{p,1P-2P} + r_e + 1)$ ODEs, instead of $(2(N_r - 1) + 3N_x + 1)$ ones as the original CSa-ESPM. Further, considering the POD truncated basis function and the solution of the (22), an approximation of the original state vectors $(\bar{c}_{s,j} \in \mathbb{R}^{N_r} \text{ and } \bar{c}_e \in \mathbb{R}^{3N_x})$ of the same dimension is reconstructed as:

$$\bar{c}_{s,n} = V_n^r \tilde{c}_{s,n} \quad \bar{c}_{s,p} = V_{p,(1P-2P)}^r \tilde{c}_{s,p} \quad \bar{c}_e = V_e^r \tilde{c}_e.$$
 (16)

The $\bar{c}_{s,j} = [\bar{c}_{s,j,1} \cdots \bar{c}_{s,j,N_r-1}]^T$ are then used to obtain an approximation of the particle surface $(\bar{c}_{s,j}^{surf} = \bar{c}_{s,j,N_r-1})$ and bulk concentration $(\bar{c}_{s,j}^{bulk} = \frac{3}{R_{s,j}^3} \int_0^{R_{s,j}} r^2 \bar{c}_{s,j} dr)$ of each

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TABLE II

POD-GALERKIN AVERAGE CORE-SHELL ESPM: SNAPSHOT MATRICES AND ROM STATE-SPACE FORMULATION





Fig. 3. Constant current discharge cycles at C/3, C/6, C/10 and C/12: Cell voltage(a), SOC (b) and moving boundary (c).

electrode. The $\bar{c}_{s,j}^{surf}$, $\bar{c}_{s,j}^{bulk}$ and \bar{c}_e^{avg} are employed, in turn, for the calculation of the approximated cell voltage (\bar{V}_{cell}), SOC (SOC_j) and as input features for the FNN, in accordance with (6–12) in Table I.

A. Simulation Results

In this section, the POD-Galerkin ROM is compared against both the high-dimensional CSa-ESPM and experimental data. The experiments are carried out on a 49.9 Ah LFP pouch cell and consist of CC discharging cycles at C/3, C/6, C/10 and C/12 rate. The high dimensional CSa-ESPM is simulated

TABLE III %RMSEs BETWEEN THE ROM AND THE CSA-ESPM

C-rate:	Discharging cycle			
	$V_{cell}^{\% RMS}$	$SOC_n^{\%RM}$	$SSOC_p^{\%RM}$	$s r_p^{\% RMS}$
C/3	0.5149	0.0066	1.1373	2.0572
C/6	0.5542	0.0194	1.3857	1.4735
C/10	0.5728	0.0959	1.2745	1.9672
C/12	0.5561	0.0066	1.4434	1.2145
	Real driving cycle			
-	0.1209	0.0023	0.0053	0.0030

considering the electrochemical parameters identified in [8] for a C/6-rate cycle and using $N_r = 70$ and $N_x = 10$ for the PDAE discretization. It should be noted that in [9], it was demonstrated that the configuration [Nr, Nx] = [70, 10]provides the best trade-off in terms of simulation accuracy. The high dimensional CSa-ESPM is employed for the snapshot matrix generation for each input value, in accordance with Section IV. The POD truncation orders used in the ROM simulation, illustrated in (23), are selected to obtain truncation degrees close to unity, as shown in (24). Finally, the comparison between both models and the experimental data, in terms of cell voltage, SOC, and moving boundary is presented in Fig. 3 and 4 for both CC discharging profiles and real driving cycles. The discrepancies between the two models are evaluated through the percentage root mean squared error (%RMSE) between Ψ^{POD} (quantity from ROM) and Ψ (quantity from the high-dimensional CSa-ESPM):

$$\Psi^{\%RMS} = \sqrt{\frac{1}{t_{end}} \int_{0}^{t_{end}} (\Psi^{POD} - \Psi)^2 dt \frac{100t_{end}}{\int_{0}^{t_{end}} \Psi dt}}$$
(25)

where $\Psi^{POD} \in [\bar{V}_{cell}, S\bar{O}C_j, \bar{r}_p]$ and $\Psi \in [V_{cell}, SOC_j, r_p]$. It is observed that the ROM matches the dynamics of the original



Fig. 4. Real driving cycle results comparison: Cell current profile (a), voltage (b) and SOC (c).

model well, as depicted in Fig. 3 and 4. In particular, it provides an accurate approximation of the cell voltage (%RMSE consistently below 0.6%) and negative SOC, as indicated by $V_{cell}^{\% RMS}$ and $SOC_n^{\% RMS}$ in Table III. In contrast, the %RMSE of positive SOC and moving boundary r_p are slightly higher, but still within 2.1%, showing that the ROM conserves the phase transition information. It needs to be stressed that the ROM results shown in Fig. 3 and 4 are obtained by solving a system of 9 ODEs (i.e., 2: negative electrode, 3+1: positive electrode + moving boundary, 3: electrolyte), instead of the 169 ODEs used for the CSa-ESPM. This stunning reduction in model-order results in low usage of memory during simulation, wherein the ROM requires only one-third of the peak memory needed by CSa-ESPM to simulate the cell dynamics for the CC profiles listed in Table III. The simulation time of the ROM, though, is in the same order of magnitude as that of CSa-ESPM, which is expected when the POD-Galerkin method is applied to reduce the order of a non-linear model [20]. Overall, the primary advantage of the POD-Galerkin ROM is the considerable reduction of the number of ODEs (from 169 to 9) describing the cell electrochemical dynamics while accounting for the phase transition, easing the computational burden, and still maintaining accuracy.

V. CONCLUSION

In this letter, a ROM is proposed to approximate the electrochemical dynamics of graphite/LFP batteries. The POD-Galerkin technique is adopted to reduce the dimensionality of the state-space model that results from the spatial discretization of the CSa-ESPM electrochemical governing equations. The resulting ROM offers a considerable dimensionality reduction - from 169 to 9 state variables. As shown from the CC and real driving cycle validation results against both experimental data and CSa-ESPM simulations, the POD-based

ROM enables an accurate match of the cell voltage and electrodes SOC. Moreover, the designed ROM is still able to capture the phase transition dynamics and accounts for the voltage hysteresis and path dependence dynamics.

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