Region-Aware Goal Programming for Electrochemical Parameter Auto-Tuning with Dynamic Critical-Point Constraints

Xu GE

UM-SJTU JI

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Outline

1. Introduction & Motivation
2. Electrochemical Modeling
3. Parameter Sensitivity Analysis
4. Problem Formulation
5. Algorithm and Implementation

Research Overview

Challenge: Accurate parameter identification for electrochemical battery models faces inherent difficulties:

- High-dimensional parameter space with complex interdependencies
- Highly nonlinear charge-discharge behavior
- Model-reality discrepancy, especially at critical states

Key Questions:

- How to capture the electrochemical dynamics accurately?
- Which parameters dominate the battery behavior?
- How to ensure robustness at critical operating points?

Motivation

Why Parameter Identification Matters:

$$\text{Model Accuracy} \propto \text{Parameter Precision} \times \text{Structure Adequacy}$$

Critical Applications:

- **Battery Management:** Real-time SoC estimation

$$\xi(t) = \xi(0) - \frac{1}{Q_n} \int_0^t \frac{\eta I(\tau)}{3600} d\tau$$

- **Life Prediction:** Capacity fade modeling

$$C_{\text{loss}} = f(\theta, \text{cycle}, T, \text{DoD})$$

- **Cell Design:** Material and structural optimization

State-of-the-Art and Limitations

Current Approaches:

- **Conventional Methods:**

$$\min_{\theta} \sum_{i=1}^{N} (V_{\text{exp}}(t_i) - V_{\text{model}}(t_i, \theta))^2$$

- **Gradient-based Optimization:**
  - Limited to local optima
  - Sensitive to initial conditions
  - Struggles with parameter coupling
- **Black-box Methods:**
  - Computationally expensive
  - Lack physical insights

Innovations

Novel Framework:

$$\begin{aligned} \min_{\theta} \quad & f(\theta) + \sum_{k} w_k e_k(\theta) \\ s.t. \quad & g_{\mathcal{L}}(\theta) \leq \epsilon_{\mathcal{L}} \quad \text{(Low SoC region)} \\ & g_{\mathcal{H}}(\theta) \leq \epsilon_{\mathcal{H}} \quad \text{(High SoC region)} \\ & g_{\mathcal{K}}(\theta) \leq \epsilon_{\mathcal{K}} \quad \text{(Key Point)} \end{aligned}$$

Key Features:

- **Multi-objective Formulation:**
  - Global curve fitting
  - Critical point accuracy
  - Region-specific constraints
- **Efficient Solution Strategy:**
  - Sensitivity-guided parameter selection
  - Scalable Bayesian optimization
  - Adaptive trust region management

Technical Roadmap

Systematic Approach:

1. **Electrochemical Modeling**
   - DFN model formulation
   - Transport phenomena characterization
2. **Parameter Analysis**
   - Sobol sensitivity indices
   - Parameter interaction analysis
3. **Optimization Framework**
   - Multi-objective formulation
   - Key point constraints
   - Region-specific error bounds

Expected Impact:

- Enhanced model accuracy in critical regions
- Reduced computational complexity
- Improved physical interpretability

DFN Model Overview

Doyle-Fuller-Newman Model:

- Multi-scale physics-based model incorporating:
  - Mass transport in electrolyte and solid phases
  - Charge conservation
  - Butler-Volmer kinetics
  - Thermodynamic equilibrium

Governing Equations - Mass Transport

Solid Phase Diffusion:

$$\frac{\partial c_s}{\partial t} = \frac{D_s}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial c_s}{\partial r}\right)$$

with boundary conditions:

$$\left.\frac{\partial c_s}{\partial r}\right|_{r=0} = 0$$

$$-D_s\left.\frac{\partial c_s}{\partial r}\right|_{r=R_s} = \frac{j_n}{a_s F}$$

Electrolyte Phase:

$$\epsilon_e\frac{\partial c_e}{\partial t} = \nabla\cdot(D_e^{\text{eff}}\nabla c_e) + \frac{1-t_+^0}{F}j_n$$

where:

- c_s: Solid phase concentration
- c_e: Electrolyte concentration
- D_s: Solid diffusion coefficient

Charge Conservation

Solid Phase Potential:

$$\nabla \cdot (\sigma^{\text{eff}} \nabla \phi_s) = j_n$$

Electrolyte Phase Potential:

$$\nabla \cdot (\kappa^{\text{eff}} \nabla \phi_e) + \nabla \cdot (\kappa_D^{\text{eff}} \nabla \ln c_e) = -j_n$$

where:

$$\sigma^{\text{eff}} = \epsilon_s^{\text{brugg}} \sigma$$
$$\kappa^{\text{eff}} = \epsilon_e^{\text{brugg}} \kappa$$
$$\kappa_D^{\text{eff}} = \frac{2RT\kappa^{\text{eff}}}{F}(t_+^0 - 1)$$

Electrochemical Kinetics

Butler-Volmer Equation:

$$j_n = i_0 \left[\exp\left(\frac{\alpha_a F}{RT} \eta \right) - \exp\left(-\frac{\alpha_c F}{RT} \eta \right) \right]$$

Exchange Current Density:

$$i_0 = k(c_e)^{\alpha_a} (c_{s,\text{max}} - c_{s,\text{surf}})^{\alpha_a} (c_{s,\text{surf}})^{\alpha_c}$$

Overpotential:

$$\eta = \phi_s - \phi_e - U(c_{s,\text{surf}})$$

where:

- k: Reaction rate constant
- α_a, α_c: Transfer coefficients
- U: Open-circuit potential

State Variables and Parameters

Key State Variables:

- Solid concentration $c_s(r, x, t)$
- Electrolyte concentration $c_e(x, t)$
- Solid potential $\phi_s(x, t)$
- Electrolyte potential $\phi_e(x, t)$

Critical Parameters for Identification:

$$\boldsymbol{\theta} = \{D_s^{\text{neg}}, D_s^{\text{pos}}, D_e, \kappa, k^{\text{neg}}, k^{\text{pos}}, \text{brugg}, \epsilon_e, \alpha_a, \alpha_c, R_s, a_s\}$$

Model Characteristics

Nonlinear Coupling:

- Multi-scale interactions
- Strong parameter interdependence
- State-dependent transport properties

Numerical Challenges:

- Stiff PDEs requiring robust solvers
- Multiple time scales
- Boundary layers at interfaces

Parameter Sensitivity:

- Varies with operating conditions
- State-dependent parameter influence
- Critical at extreme states (high/low SoC)

Global Sensitivity Analysis Overview

Motivation:

- Curse of dimensionality in parameter space
- Complex parameter interactions
- Computational efficiency requirements

Sobol Method Advantages:

- Captures nonlinear effects
- Quantifies parameter interactions
- Model-independent approach
- Variance-based decomposition

Sobol Decomposition Theory

ANOVA-HDMR Decomposition:

$$V_{\text{model}}(t_i) = f_0 + \sum_{i=1}^{d} f_i(\theta_i) + \sum_{i<j}^{d} f_{ij}(\theta_i, \theta_j) + \cdots + f_{1,2,\ldots,d}(\theta_1, \ldots, \theta_d)$$

Total Variance Decomposition:

$$V(Y) = \sum_{i=1}^{d} V_i + \sum_{i<j}^{d} V_{ij} + \cdots + V_{1,2,\ldots,d}$$

where:

- V_i: Individual parameter variance
- V_{ij}: Second-order interaction variance
- Higher-order terms represent complex interactions

Sobol Sensitivity Indices

First-order Indices:

$$S_i = \frac{V_i}{V(Y)} = \frac{V_{\theta_i}(\mathbb{E}_{\theta_{\sim i}}[Y|\theta_i])}{V(Y)}$$

Total Effect Indices:

$$ST_i = 1 - \frac{V_{\theta_{\sim i}}(\mathbb{E}_{\theta_i}[Y|\theta_{\sim i}])}{V(Y)}$$

Interpretation:

- S_i: Direct influence of parameter i
- $ST_i - S_i$: Interaction effects
- $\sum_i S_i \leq 1$: Model nonlinearity measure

Monte Carlo Estimation

Numerical Computation:

$$\hat{S}_i = \frac{\frac{1}{N}\sum_{j=1}^{N} f(A)_j[f(B_A^i)_j - f(B)_j]}{\frac{1}{N}\sum_{j=1}^{N} f(A)_j^2 - f_0^2}$$

Implementation:

- Two independent sampling matrices (A, B)
- Resampling strategy for variance estimation
- Convergence analysis
- Bootstrap confidence intervals

Computational Cost:

$$N_{\text{total}} = N(k+2) \text{ model evaluations}$$

Analysis Results

Parameter Ranking:

Parameter	First-order	Total Effect	Rank
D_s^{pos}	0.42	0.48	1
k^{pos}	0.25	0.32	2
D_e	0.15	0.22	3
ϵ_e	0.08	0.12	4
$\vdots$	$\vdots$	$\vdots$	$\vdots$

Implications for Parameter Identification

Reduced Parameter Set:

$$\boldsymbol{\theta}_{\text{reduced}} = \{\theta_i | ST_i > \text{threshold}\}$$

Benefits:

- Dimensionality reduction: $d_{\text{reduced}} \ll d_{\text{original}}$
- Improved optimization convergence
- Enhanced parameter identifiability
- Reduced computational cost

Validation Strategy:

- Cross-validation using different operating conditions
- Error analysis on reduced parameter set
- Robustness assessment

Problem Structure Overview

Key Challenges in Parameter Identification:

- Global curve fitting accuracy
- Critical point precision
- Region-specific performance

Multi-objective Formulation:

$$\min_{\boldsymbol{\theta}} \{f(\boldsymbol{\theta}),\, g_{\mathcal{K}}(\boldsymbol{\theta}),\, g_{\mathcal{L}}(\boldsymbol{\theta}),\, g_{\mathcal{H}}(\boldsymbol{\theta})\}$$

where:

- $f(\boldsymbol{\theta})$: Global fitting objective
- $g_{\mathcal{K}}(\boldsymbol{\theta})$: Key point constraints
- $g_{\mathcal{L}}(\boldsymbol{\theta}), g_{\mathcal{H}}(\boldsymbol{\theta})$: Region-specific constraints

Global Fitting Objective

Root Mean Square Error:

$$f(\boldsymbol{\theta}) = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(\hat{V}(t_i;\boldsymbol{\theta}) - V(t_i)\right)^2}$$

Parameter Bounds:

$$\boldsymbol{\theta}_{\mathsf{min}} \leq \boldsymbol{\theta} \leq \boldsymbol{\theta}_{\mathsf{max}}$$

Physical Constraints:

$$\begin{aligned} c_s &\leq c_{s,\mathsf{max}} \\ 0 &\leq c_e \\ \epsilon_s + \epsilon_e &\leq 1 \end{aligned}$$

Key Points Identification

Categories of Key Points:

- **Inflection Points:**

$$\mathcal{K}_{\text{inf}} = \left\{ t_k \middle| \left(\frac{d^2V}{dt^2} \right)_{t=t_k} = 0 \right\}$$

- **Phase Transitions:**

$$\mathcal{K}_{\text{phase}} = \left\{ t_k \middle| \left(\frac{dV}{dt} \right)_{t=t_k} > \delta_{\text{phase}} \right\}$$

- **Capacity Points:**

$$\mathcal{K}_{\text{cap}} = \{ t_k | \xi(t_k) \in \{0.2, 0.5, 0.8\} \}$$

Complete Key Point Set:

$$\mathcal{K} = \mathcal{K}_{\text{inf}} \cup \mathcal{K}_{\text{phase}} \cup \mathcal{K}_{\text{cap}}$$

Inflection Points Detection

Numerical Derivative Computation:

$$\frac{dV}{dt} \approx \frac{V(t_{i+1}) - V(t_{i-1})}{2\Delta t}$$
$$\frac{d^2V}{dt^2} \approx \frac{V(t_{i+1}) - 2V(t_i) + V(t_{i-1})}{\Delta t^2}$$

Robust Detection Strategy:

1. Apply Savitzky-Golay filter for noise reduction:

$$V_{\text{smooth}}(t_i) = \sum_{j=-n}^{n} c_j V(t_{i+j})$$

2. Compute derivatives on smoothed data
3. Zero-crossing detection for second derivative:

$$\text{sign}\left(\frac{d^2V_{\text{smooth}}}{dt^2}\right)_{t_i} \neq \text{sign}\left(\frac{d^2V_{\text{smooth}}}{dt^2}\right)_{t_{i+1}}$$

Adaptive Weight Assignment

Curvature-Based Weights:

$$\kappa(t_k) = \frac{\left|\frac{d^2V}{dt^2}\right|_{t=t_k}}{\left(1 + \left(\frac{dV}{dt}\right)^2_{t=t_k}\right)^{3/2}}$$

Local Information Content:

$$I(t_k) = -\ln\left(\frac{\sigma_k^2}{\sigma^2_{\max}}\right)$$

where σ_k^2 is local variance in neighborhood of t_k

Final Weight Computation: $w_k = \alpha_k \exp(-\beta_k \kappa(t_k)) \cdot \exp(\gamma_k I(t_k))$

Parameter Selection:

- α_k: Point type importance
- β_k: Curvature sensitivity
- γ_k: Information weight

Region-Specific Constraints

State of Charge Definition: $\xi(t) := \mathrm{SoC}(t) \in [0, 1]$
Low SoC Region ($\mathcal{L}$):

$$g_{\mathcal{L}}(\boldsymbol{\theta}) = \max_{t_i \in \mathcal{T}_{\mathcal{L}}} |\hat{V}(t_i; \boldsymbol{\theta}) - V(t_i)|$$
$$g_{\mathcal{L}}(\boldsymbol{\theta}) - d^{+}_{\mathcal{L},t_i} + d^{-}_{\mathcal{L},t_i} = \epsilon_{\mathcal{L}}, \quad d^{+}_{\mathcal{L},t_i}, d^{-}_{\mathcal{L},t_i} \geq 0, \quad \forall t_i \in \mathcal{T}_{\mathcal{L}}$$

High SoC Region ($\mathcal{H}$):

$$g_{\mathcal{H}}(\boldsymbol{\theta}) = \max_{t_i \in \mathcal{T}_{\mathcal{H}}} |\hat{V}(t_i; \boldsymbol{\theta}) - V(t_i)|$$
$$g_{\mathcal{H}}(\boldsymbol{\theta}) - d^{+}_{\mathcal{H},t_i} + d^{-}_{\mathcal{H},t_i} = \epsilon_{\mathcal{H}}, \quad d^{+}_{\mathcal{H},t_i}, d^{-}_{\mathcal{H},t_i} \geq 0, \quad \forall t_i \in \mathcal{T}_{\mathcal{H}}$$

Region-Specific Constraints

Key Point Constraint ($\mathcal{K}$):

$$g_{\mathcal{K}}(\boldsymbol{\theta}) = \max_{t_i \in \mathcal{T}_{\mathcal{K}}} |\hat{V}(t_i; \boldsymbol{\theta}) - V(t_i)|$$

$$g_{\mathcal{K}}(\boldsymbol{\theta}) - d^{+}_{\mathcal{K},t_i} + d^{-}_{\mathcal{K},t_i} = \epsilon_{\mathcal{K}}, \quad d^{+}_{\mathcal{K},t_i}, d^{-}_{\mathcal{K},t_i} \geq 0, \quad \forall t_i \in \mathcal{T}_{\mathcal{K}}$$

Region Definition:

$$\mathcal{T}_{\mathcal{L}} = \{t_i \mid \xi(t_i) \leq \xi_{\mathcal{L}}\}$$
$$\mathcal{T}_{\mathcal{H}} = \{t_i \mid \xi(t_i) \geq \xi_{\mathcal{H}}\}$$
$$\mathcal{T}_{\mathcal{K}} = \{t_i \mid t_i \text{ corresponds to key points}\}$$

Goal Programming Formulation

Objective Function:

$$\min_{\boldsymbol{\theta}} \quad w_1 f(\boldsymbol{\theta}) + w_2 \sum_{t_i \in \mathcal{T}_\mathcal{K}} d^+_{\mathcal{K},t_i} + w_3 \sum_{t_i \in \mathcal{T}_\mathcal{L}} d^+_{\mathcal{L},t_i} + w_4 \sum_{t_i \in \mathcal{T}_\mathcal{H}} d^+_{\mathcal{H},t_i}$$

Constraints:

$$\begin{aligned} g_\mathcal{L}(\boldsymbol{\theta}) - d^+_{\mathcal{L},t_i} + d^-_{\mathcal{L},t_i} &= \epsilon_\mathcal{L}, \quad \forall t_i \in \mathcal{T}_\mathcal{L} \\ g_\mathcal{H}(\boldsymbol{\theta}) - d^+_{\mathcal{H},t_i} + d^-_{\mathcal{H},t_i} &= \epsilon_\mathcal{H}, \quad \forall t_i \in \mathcal{T}_\mathcal{H} \\ g_\mathcal{K}(\boldsymbol{\theta}) - d^+_{\mathcal{K},t_i} + d^-_{\mathcal{K},t_i} &= \epsilon_\mathcal{K}, \quad \forall t_i \in \mathcal{T}_\mathcal{K} \\ d^+_{\mathcal{L},t_i}, d^-_{\mathcal{L},t_i}, d^+_{\mathcal{H},t_i}, d^-_{\mathcal{H},t_i}, d^+_{\mathcal{K},t_i}, d^-_{\mathcal{K},t_i} &\geq 0, \quad \forall t_i \end{aligned}$$

Weight Selection:

- w_1: Global fit priority
- w_2: Key point importance
- w_3: Low SoC region-specific weights
- w_4: High SoC region-specific weights

Bayesian Optimization Framework

Core Components and Workflow:

- **Gaussian Process Surrogate Model:** Efficiently models the objective and constraints with uncertainty quantification.
- **Trust Region Strategy:** Guides local exploration and ensures convergence.
- **Thompson Sampling:** Balances exploration and exploitation for candidate selection.
- **Batch Evaluations:** Leverages parallel resources for efficiency.

Workflow Overview:

Sample Candidates $\rightarrow$ Update Surrogate Models $\rightarrow$ Optimize Acquisition Function $\rightarrow$ Evaluate Candidates $\rightarrow$ Adapt Trust Region

Surrogate Model Design

Gaussian Process Surrogate Models:

- **Objective Function:**

$$f(\boldsymbol{\theta}) \sim \mathcal{GP}(m_f(\boldsymbol{\theta}), k_f(\boldsymbol{\theta}, \boldsymbol{\theta}'))$$

- **Constraint Functions:**

$$g_i(\boldsymbol{\theta}) \sim \mathcal{GP}(m_i(\boldsymbol{\theta}), k_i(\boldsymbol{\theta}, \boldsymbol{\theta}'))$$

Kernel Function:

$$k(\boldsymbol{\theta}, \boldsymbol{\theta}') = \sigma^2 \prod_{i=1}^{d} \exp\left(-\frac{(\theta_i - \theta_i')^2}{2l_i^2}\right)$$

Key Properties: Scalable, differentiable, and suitable for high-dimensional spaces.

Trust Region and Candidate Selection

Trust Region Management:

- **Region Definition:** $\text{TR} = \{\boldsymbol{\theta} : \|\boldsymbol{\theta} - \boldsymbol{\theta}_c\|_\infty \leq L\}$
- **Adaptive Updates:**
  - **Success:** $L_{\text{new}} = \min\{2L, L_{\text{max}}\}$
  - **Failure:** $L_{\text{new}} = L/2$
  - **Restart:** Triggered if $L < L_{\text{min}}$.

Thompson Sampling for Candidate Selection:

- Generate candidates $\{\boldsymbol{\theta}_i\}_{i=1}^{q}$ using low-discrepancy Sobol sequences within TR.
- Evaluate posterior realizations for both objective and constraints:

$$\tilde{f}_i \sim \mathcal{GP}(m_f, k_f)|\boldsymbol{\theta}_i$$
$$\tilde{g}_i \sim \mathcal{GP}(m_g, k_g)|\boldsymbol{\theta}_i$$

- Select the most promising candidate:

$$\boldsymbol{\theta}^* = \underset{\boldsymbol{\theta}_i \in \mathcal{F}_{\text{sample}}}{\arg\min}\ \tilde{f}_i \quad \text{s.t.} \quad \tilde{g}_i \leq 0.$$

Batch Evaluation Strategy and Implementation

Batch Construction:

- Select initial candidate:

$$\boldsymbol{\theta}_1 = \arg\min_{\boldsymbol{\theta}} \mathbb{E}[I(\boldsymbol{\theta})].$$

- Ensure diversity by maximizing pairwise distance:

$$\boldsymbol{\theta}_k = \arg\max_{\boldsymbol{\theta}} \min_{i<k} \|\boldsymbol{\theta} - \boldsymbol{\theta}_i\|_2.$$

Parallel Execution:

- Evaluate q candidates simultaneously.
- Update surrogate models asynchronously to incorporate feedback.

Implementation Details:

- GPU acceleration for Gaussian process regression.
- Scalable numerical solvers for kernel matrix inversion.
- Hyperparameter tuning for initial trust region size L_{init} and convergence criteria.