**Region-Aware Cross-Condition Electrochemical Parameter Identification of High-Capacity Lithium-Ion Battery Through Sequential Global-Local Optimization**

**1. Introduction**

* **Background**: The identification of electrochemical parameters in lithium-ion batteries (LIBs) is crucial for optimizing battery performance, particularly for high-capacity cells (>200Ah). This section elaborates on the current challenges of parameter identification, especially for large cells, and their impact on battery modeling accuracy and reliability.
* **Literature Review**:
  + **Physical Experiment Methods**: Discuss invasive techniques like Ecker et al. (2008) and Schmalstieg et al. (2018) that require cell disassembly, highlighting their limitations in accuracy, expense, and time consumption.
  + **Gradient-Based Methods**: Review methods like Levenberg-Marquardt (Santhanagopalan et al., 2008) and Gauss-Newton, emphasizing their susceptibility to local minima in the complex parameter landscape.
  + **Meta-heuristic Approaches**: Critically assess Genetic Algorithms (Forman et al., 2012), Particle Swarm Optimization (Rahman et al., 2016), and Cuckoo Search, noting their computational expense and convergence issues for large parameter sets.
  + **Bayesian Methods**: Review recent work on Bayesian parameter identification (Kim et al., 2023), highlighting limitations in handling multiple C-rate conditions simultaneously.
  + **Data-Driven Optimization**: Analyze frameworks like those proposed by Lai et al. (2021), focusing on their limitations for high-capacity cells.
* **Research Gap**: Despite advances in parameter identification methods, there remains a significant gap in methodologies specifically tailored for high-capacity batteries (>200Ah), which exhibit unique electrochemical behaviors. Current approaches fail to achieve consistent accuracy across multiple C-rates and lack efficient convergence for the complex parameter spaces of these large cells.
* **Innovation Points**:
  + First comprehensive parameter identification framework specifically for high-capacity (280Ah) LIBs
  + Novel cross-condition min-max optimization approach ensuring robustness across multiple discharge rates
  + Region-aware sequential global-local optimization that strategically transitions between exploration and exploitation
* **Paper Structure**: Expanded outline of subsequent sections with emphasis on methodological innovations
* **为什么要进行贝叶斯优化与局部最优的协同：首先贝叶斯优化不能保证最后迭代算出解的局部最优性，所以需要加一个局部最优的local算法确保局部最优。**

**2. Doyle-Fuller-Newman (DFN) Model Development**

* **Theoretical Foundation**: Comprehensive presentation of the governing equations of the DFN model with specific emphasis on adaptations required for high-capacity cells
  + Mass conservation equations with consideration of large electrode thickness effects
  + Modified charge conservation equations accounting for high-capacity cell-specific phenomena
  + Enhanced Butler-Volmer kinetics formulation for high-current applications
  + Comprehensive thermal modeling crucial for large-format cells
* **Model Implementation**: Detailed implementation using the PyBaMM framework with custom modifications for high-capacity cells
* **Boundary and Initial Conditions**: Specialized formulations for different discharge rates with particular attention to boundary effects in large cells
* **Parameters Categorization**: Systematic classification of parameters into electrochemical, transport, geometric, and thermal groups, highlighting their physical significance and interdependencies in high-capacity cells

**3. Sensitivity Analysis**

* **Advanced Sensitivity Framework**: Implementation of global Sobol sensitivity analysis with special attention to computational efficiency for high-dimensional parameter spaces
* **Multi-rate Sensitivity Analysis**: Novel approach examining parameter sensitivity across different C-rates simultaneously
* **Parameter Interaction Analysis**: Investigation of parameter interactions and their impact on model outputs
* **Sensitivity-Based Parameter Prioritization**: Development of a hierarchical parameter selection strategy based on cross-rate sensitivity significance
* **Sensitivity Visualization**: Advanced visualization techniques to interpret complex sensitivity relationships
* **Cross-Condition Sensitivity Dynamics**: Analysis of how sensitivity patterns shift with changing discharge conditions, with implications for robust parameter identification

**4. Region-Aware Min-Max Optimization Algorithm**

* **Problem Formulation**:
  + Development of a multi-objective function that minimizes RMSE across different C-rates (0.1C 0.2C 0.33C 1C) simultaneously
  + Introduction of novel min-max optimization formulation to ensure cross-condition robustness (minimize the Max RMSE under four condition)
  + Add hard constraints to key regions of the discharge curve, such as adding hard constraints with RMSE less than 10mv in the low SOC (0-20% SOC) and high SOC segments (80-100% SOC)
* **Bayesian Optimization Framework**: Scalable Constrained Bayesian Optimization
  + Advanced Gaussian process surrogate modeling with specialized kernels for electrochemical systems
  + Multi-point acquisition function strategy to enhance exploration efficiency
  + Batch optimization techniques to leverage parallel computing resources
* **Local Refinement**:
  + Adaptive interior-point method with path-following techniques
  + Modified sequential quadratic programming with trust-region strategies
  + Specialized convergence criteria for electrochemical parameter spaces
* **Global-Local Synergistic Approach**:
  + Dynamic transition mechanism between global and local search phases
  + Region-aware strategy that adapts search space based on parameter sensitivity and uncertainty
  + Adaptive trust region definition for efficient parameter space exploration
  + Comprehensive algorithm workflow with built-in uncertainty quantification

**5. Experimental Setup**

* **Cell Specifications**: Detailed characterization of the 280Ah lithium-ion battery including electrode composition, cell geometry, and manufacturer specifications
* **Advanced Testing Infrastructure**: Description of high-precision testing equipment capable of handling high-capacity cells
* **Comprehensive Experimental Protocol**:
  + Systematic discharge tests at various C-rates (0.1C, 0.2C, 0.33C, 1C)
  + Precision temperature control and monitoring systems
  + Statistical design of experiments for robust validation

# 6. Experimental Results and Discussion

## 6.1 Parameter Identification Results

* **Table 1**: Comparison of identified parameters using different optimization strategies
  + Standard Bayesian optimization
  + Bayesian optimization with local refinement
  + Constrained Bayesian optimization with local refinement
* Analysis of parameter differences and physical significance
* Discussion on how parameter constraints improve identification accuracy

## 6.2 Discharge Performance Validation at Multiple C-rates

* **Figure 1**: Validation of the proposed method at multiple discharge rates
  + Four subplots showing actual vs. simulated discharge curves for battery #81 at 0.1C, 0.2C, 0.33C, and 1C rates
  + Voltage error analysis for each C-rate
* **Figure 2**: Cross-cell validation
  + Comparison of discharge curves for batteries #81 and #82 at all four C-rates in a single plot
  + Magnified view of critical discharge regions showing model accuracy

## 6.3 Comparative Analysis of Optimization Strategies

* **Figure 3**: Performance comparison of different optimization approaches
  + Discharge curve comparison between standard Bayesian optimization, Bayesian with local refinement, and constrained Bayesian with local refinement
  + Analysis of error reduction achieved by the proposed method across different C-rates
* Discussion on the advantages of the region-aware cross-condition approach

## 6.4 Model Validation Under Random Current Profiles

* **Figure 4**: Performance validation under dynamic operating conditions
  + Comparison between measured and simulated voltage responses under random current profiles
  + Error distribution analysis
* Discussion on model robustness for real-world applications

## 6.5 Computational Efficiency Analysis

* **Table 2**: Comparison of computational times
  + Execution time for each optimization strategy
  + Trade-off between computational cost and model accuracy
* Discussion on the practical implementation considerations

**7. Conclusion and Future Work**

* **Key Findings**: Synthesis of main results highlighting the superior performance of the proposed methodology
* **Scientific and Practical Contributions**:
  + First comprehensive parameter identification framework for high-capacity (280Ah) cells with demonstrated cross-rate robustness
  + Novel min-max optimization approach that ensures consistent accuracy across multiple operating conditions
  + Methodological innovation in the integration of global Bayesian search with region-aware local refinement
  + Practical implications for battery management systems and thermal management strategies
* **Limitations and Challenges**: Critical assessment of the current approach, including computational complexity and experimental constraints
* **Future Research Directions**:
  + Extension to other cell chemistries and form factors
  + Integration of aging mechanisms in the parameter identification framework
  + Real-time implementation strategies for battery management systems
  + Multi-cell and pack-level parameter identification considerations