Battery Design Optimization using PyBaMM and Bayesian Optimization

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Abstract. This project applies Bayesian Optimization (BO) to optimize lithium-ion battery design parameters using PyBaMM, a physics-based electrochemical model. The approach replaces gradient-based search with a Gaussian Process surrogate to efficiently maximize energy density while maintaining voltage and manufacturability constraints. Results demonstrate improved energy density and reveal intrinsic trade-offs between energy, power, and durability, highlighting the potential of Bayesian Optimization as a robust framework for automated battery design.

Keywords: Bayesian Optimization · PyBaMM · lithium-ion batteries · electrode design · energy density · Gaussian Process

1 Introduction

Lithium-ion battery design involves a complex multi-objective optimization problem. Parameters such as electrode thickness, porosity, and particle size strongly influence energy density, power density, thermal behavior, and lifetime. Therefore, finding the right trade-off between these competing objectives is essential for the development of advanced battery systems.

Given this motivation, this project focuses on applying **Bayesian Optimization (BO)** to a constrained battery design problem, coupled with physics-based battery modeling using **PyBaMM**, in order to automatically identify optimal design parameters that maximize energy density while satisfying operational and manufacturing constraints.

2 Project Overview

The project builds upon an initial implementation using deterministic optimization (via Scipy's SLSQP algorithm) and extends it using Bayesian Optimization to explore the design space more efficiently.

The study is based on the Doyle–Fuller–Newman (DFN) model [1] implemented in **PyBaMM** (Python Battery Mathematical Modelling). This model captures the electrochemical and transport phenomena inside lithium-ion cells, enabling a realistic prediction of voltage response, current, and energy output under different design configurations.

Although the first optimization approach implemented in this work is fundamentally a gradient-based optimization method, the objective function is derived from a PyBaMM battery simulation, which represents a black-box process with no analytical expression. In such cases, numerical gradient estimation can be computationally expensive and potentially unstable, leading to unreliable convergence. For this reason, the **Bayesian Optimization** approach provides a more efficient and robust alternative, as it does not rely on derivative information and is better suited for optimizing black-box and computationally expensive functions.

Optimization Objective

The optimization seeks to maximize the cell's energy density E_d, defined as:

$$MaximizeE_d = \frac{E}{V} = \frac{1}{3600} \frac{\int_0^T V(t)I(t)\,dt}{A \cdot L} \tag{1}$$

subject to:

- $V_{min} \geq 3.0\,V$
- $L_p + L_n \leq 20 \times 10^{-5}\,m$
- $0.2 \leq \varepsilon_p \leq 0.4$

where:

- L_p, L_n are the positive and negative electrode thicknesses,
- ε_p is the positive electrode porosity,
- A is the cell area (assumed $1\,m^2$).

3 Theoretical Background

3.1 Battery Modeling with PyBaMM

PyBaMM is an open-source Python framework that implements physics-based electrochemical battery models [2]. It enables simulation of the DFN model, the SPM (Single Particle Model), and other reduced-order models. The DFN model resolves lithium-ion concentration gradients across electrodes and electrolyte, capturing effects such as overpotentials, solid diffusion, and electrolyte polarization—factors that are critical to realistic energy density predictions.

3.2 Influence of Electrode Parameters

Battery performance is strongly dependent on several design variables that govern electrochemical transport and reaction dynamics. Variations in electrode thickness, porosity, and particle size directly affect both energy density and power capability, introducing key trade-offs that must be balanced in the optimization process.

Table 1. Influence of electrode design parameters on battery performance.

Parameter	Effect on Energy	Effect on Power
↑ Electrode thickness	↑ Energy (more active material)	↓ Power (higher resistance)
↓ Porosity	↑ Energy (denser active material)	↓ Power (slower ion transport)
↑ Particle radius	↓ Surface area (less reaction rate)	↓ Power capability
↑ Separator thickness	No direct effect	↓ Power (increased ionic resistance, longer ion path)

Electrode thickness refers to the physical height of the active material layer coated onto the current collector, typically measured in micrometers (μm), while electrode porosity is defined as the volume fraction of void space within this electrode structure, filled with liquid electrolyte that facilitates lithium-ion transport. A porosity of 0.3, for example, indicates that 30% of the electrode volume consists of electrolyte-filled pores, while the remaining 70% comprises solid active material, binders, and conductive additives.

Increasing electrode thickness generally enhances the cell's energy density by providing more active material for lithium intercalation. However, this also lengthens the ion transport path within both the electrode and the electrolyte, leading to higher internal resistance and a larger voltage drop during operation. Consequently, thicker electrodes can store more energy but deliver less power, particularly at high discharge rates.

Similarly, electrode porosity and particle size strongly influence ionic and electronic transport. Lower porosity increases the volume fraction of active material, improving theoretical energy density, but it simultaneously restricts the diffusion of lithium ions through the electrolyte-filled pores, reducing both power capability and rate performance. Therefore, optimal electrode design requires balancing energy density, voltage stability, and transport efficiency.

3.3 Bayesian Optimization

Unlike gradient-based methods, **Bayesian Optimization (BO)** efficiently explores high-dimensional design spaces with computationally expensive objective functions. It constructs a Gaussian Process (GP) surrogate model to approximate the objective function and employs an acquisition function to select the most informative next point to evaluate. This strategy enables a significant reduction in the number of **PyBaMM** simulations required to converge to an optimal design, making BO particularly suitable for physics-based battery modeling where each evaluation involves high computational cost.

4 Methodology

4.1 Base Model and Deterministic Optimization

A deterministic optimization was first performed to identify the influence of key design variables—namely the positive and negative electrode thicknesses—on the cell's energy density. The implementation uses the SLSQP method from `scipy.optimize` with bounded variables:

```
positive_thickness = x[0]
negative_thickness = x[1]

params_dict = {
    "Positive electrode thickness [m]": positive_thickness,
    "Negative electrode thickness [m]": negative_thickness
}
results = self.simulate_battery(params_dict)
return -results["energy_density"]
```

The constraints ensure that the voltage never drops below 3 V, and the total electrode thickness remains within manufacturing limits (below 20µm). The search space was restricted to physically feasible ranges:

```
bounds = [(5.0e-5, 12.0e-5), (6.0e-5, 12.0e-5)]
result = minimize(objective_function, initial_guess, bounds=bounds)
```

Constraint violations were penalized by assigning a large negative objective value (10 Wh/L) whenever the minimum voltage dropped below 3 V or the total thickness exceeded limits.

This deterministic baseline served as a reference for the subsequent Bayesian Optimization stage, where additional design parameters—porosity, particle radius, and separator thickness—were introduced to explore higher-dimensional design trade-offs.

4.2 Bayesian Optimization Framework

The Bayesian version replaces the SLSQP loop with a **GPyOpt** [3] Bayesian optimizer. The **PyBOP** [4] library, specifically designed for battery optimization, was also considered. However, due to its recent release (2023–2024) and limited documentation, **GPyOpt** was selected as a more mature and general-purpose framework.

The design variables initially included only the positive and negative electrode thicknesses, which were optimized using Bayesian Optimization with the Maximum Probability of Improvement (MPI) acquisition function. The model was then extended to incorporate separator thickness, electrode porosity, and particle radius, resulting in a seven-dimensional optimization problem:

```
domain = [
    {'name': 'pos_thk', 'type': 'continuous', 'domain': (5e-5, 12e-5)},
    {'name': 'neg_thk', 'type': 'continuous', 'domain': (6e-5, 12e-5)},
    {'name': 'sep_thk', 'type': 'continuous', 'domain': (1e-5, 3e-5)},
    {'name': 'pos_eps', 'type': 'continuous', 'domain': (0.25, 0.40)},
    {'name': 'neg_eps', 'type': 'continuous', 'domain': (0.25, 0.40)},
    {'name': 'pos_rad', 'type': 'continuous', 'domain': (1e-6, 8e-6)},
    {'name': 'neg_rad', 'type': 'continuous', 'domain': (1e-6, 1e-5)},
]
```

Two additional parameters were introduced in this extended model, apart from the ones recommended, separator thickness and negative particle radius

The *separator thickness* (L_s) influences ionic transport resistance within the cell, as a thicker separator increases the effective diffusion path for lithium ions, reducing power capability and slightly affecting the achievable energy density. Although the separator does not store charge, its thickness determines the internal electrolyte impedance and thermal stability margin. Typical commercial values range between 10–30 μm, as reported by Ecker *et al.* (2015) and Safari and Delacourt (2011), so the design space was set to $(1 \times 10^{-5}, 3 \times 10^{-5})$ m (10–30 μm).

The *negative particle radius* (R_n) was additionally introduced to capture its influence on lithium diffusion kinetics within the solid phase. Smaller particles improve rate capability by shortening diffusion distances but can increase surface reactivity and degradation, while larger particles enhance volumetric energy density. Typical values used in DFN parameterizations, such as the Chen2020 dataset [5] and the Ecker2015 NMC cell [6], range from 1–10 μm; hence, the optimization bounds were chosen as $(1\times10^{-6}, 1\times10^{-5})$ m for the negative electrode. The positive particle radius (R_p) was already part of the initial formulation and retained the same physical range.

These two parameters were included to better capture the trade-off between ionic transport in the separator and solid-phase diffusion within the active materials—factors not represented in the initial two-variable formulation but critical for realistic multi-physics battery design.

While all tested acquisition functions—Expected Improvement (EI), Lower Confidence Bound (LCB), and MPI—converged to feasible solutions, the MPI consistently yielded higher energy density values for the same number of iterations.

```
# Objective: maximize energy density via Bayesian Optimization
kernel = GPy.kern.Matern52(input_dim=len(domain), ARD=True)
bo = GPyOpt.methods.BayesianOptimization(
    f=f, domain=domain, kernel=kernel,
    acquisition_type='MPI', acquisition_jitter=0.01
)
bo.run_optimization(max_iter=20, verbosity=True)
```

Bayesian Optimization effectively identified configurations with improved energy density while exposing intrinsic trade-offs among competing performance metrics. These relationships are discussed in the following section.

5 Design Trade-offs

From a physical perspective, the optimization problem reveals fundamental trade-offs in electrode design:

- **Energy vs. Power:** Thicker electrodes store more energy but increase internal resistance, reducing power capability.
- **Energy vs. Durability:** High-energy designs often result in stronger concentration gradients, which accelerate degradation and reduce cycle life.
- **Manufacturability:** Industrial coating processes impose limits on electrode thickness uniformity and porosity range, constraining the feasible design space.

These relationships emphasize the need for **multi-objective optimization** to simultaneously balance energy density, power density, and degradation behavior in real-world battery designs.

6 Results and Discussion

The Bayesian Optimization framework effectively identified an improved configuration that increased the cell's volumetric energy density while preserving voltage stability and manufacturability constraints. Over 55 iterations, the Gaussian Process model guided exploration across the seven-dimensional parameter space, efficiently rejecting infeasible points and refining promising regions.

The optimized configuration achieved an energy density of **110.48 Wh/L**, compared to the baseline value of **103.32 Wh/L**, representing a relative improvement of approximately **6.9%**. Table 2 summarizes the final parameter set obtained after convergence.

Table 2. Final optimized parameters obtained through Bayesian Optimization.

Parameter	**Symbol**	**Optimized Value**
Positive electrode thickness	L_p	7.45×10^{-5} m (74.47 µm)
Negative electrode thickness	L_n	1.17×10^{-4} m (117.09 µm)
Separator thickness	L_s	1.01×10^{-5} m (10.13 µm)
Positive electrode porosity	ε_p	0.3869
Negative electrode porosity	ε_n	0.3480
Positive particle radius	R_p	2.33×10^{-6} m (2.33 µm)
Negative particle radius	R_n	9.60×10^{-6} m (9.60 µm)

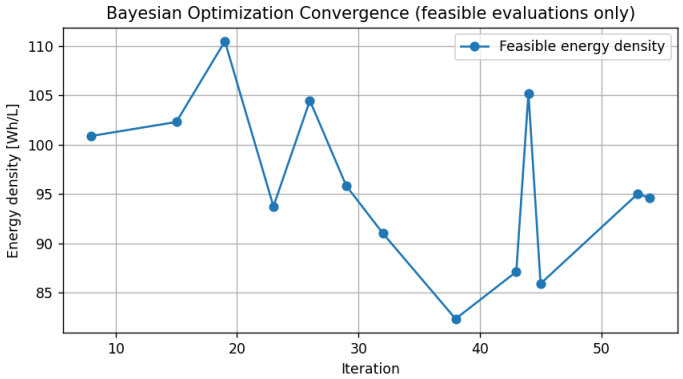

Fig. 1. Bayesian Optimization convergence showing the evaluated feasible energy densities over 55 iterations. The curve displays only non-penalized runs (with $V_{min} \geq 3.0$ V).

The total electrode stack thickness was **201.69 µm**, remaining within feasible manufacturing limits. The optimized design features thicker electrodes and larger negative particles, enabling higher active mass utilization and reduced polarization during discharge, as seen in Figure 2.

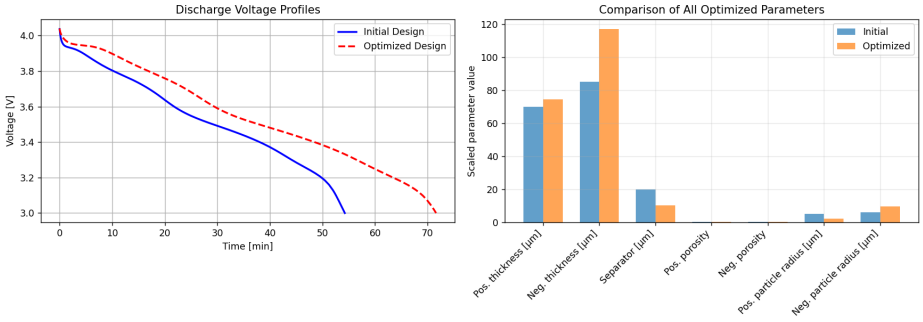

Fig. 2. (Left) Discharge voltage profiles comparing the initial and optimized configurations. (Right) Normalized parameter values before and after optimization. The optimized cell sustains higher voltage under load and longer discharge duration.

Figure 3 reports the relative importance obtained from the inverse squared ARD lengthscales of the Gaussian Process. The most influential variable in this run was the **negative electrode particle radius** (R_n), followed by the **negative electrode thickness** (L_n) and the **separator thickness** (L_s). These three parameters together account for the majority of the explained variance in the surrogate model. The **positive electrode thickness** (L_p) had a smaller but still noticeable effect, whereas porosity variables (ε_p, ε_n) and the positive

particle radius (R_p) showed very low sensitivity within the explored ranges. This indicates that, for the DFN setup and constraints used here, transport limitations on the negative side and through the separator dominated the achievable energy density.

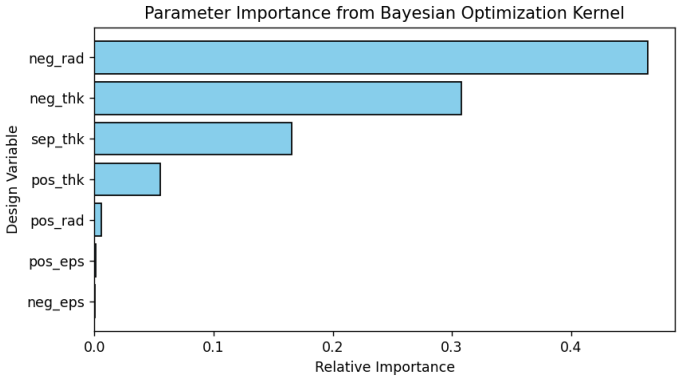

Fig. 3. Relative importance of design parameters estimated from the Gaussian Process kernel lengthscales. Positive electrode porosity shows the strongest sensitivity to changes in energy density.

Overall, the results confirm that Bayesian Optimization constitutes a robust and sample-efficient framework for physics-based battery design. In the present configuration, the optimization process revealed that the achievable energy density is primarily governed by parameters controlling lithium transport and storage within the negative electrode domain—namely the *negative particle radius*, *negative electrode thickness*, and *separator thickness*. These parameters collectively determine the effective diffusion pathways and ionic resistance, which directly influence the discharge voltage profile and total extractable energy. Conversely, variations in porosity and positive-side parameters exhibited marginal influence within the explored range. This outcome highlights that, under the DFN model and manufacturing constraints considered, **solid-phase diffusion and separator resistance represent the dominant design levers for improving volumetric energy density**.

References

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