

An optimum design methodology for battery component structures
using topology optimization

Naoyuki Ishida

KYOTO UNIVERSITY

DISSERTATION

**An optimum design methodology for battery component structures
using topology optimization**

Author:

Naoyuki Ishida

Advisor:

Professor Kazuhiro Izui

Committee member:

Professor Hiroshi Iwai

Professor Atsushi Matsubara

Professor Masashi Kishimoto

*A thesis submitted in fulfillment of the requirements
for the Doctor of Engineering*

of the

Department of Mechanical Engineering and Science
Strength of Materials and Structures Lab
Graduate School of Engineering

December 18, 2025

Declaration of Authorship

I, Naoyuki Ishida, declare that this thesis titled, “An optimum design methodology for battery component structures using topology optimization” and the work presented in it are my own. I confirm that:

- This work was done wholly or mainly while in candidature for a research degree at this University.
- Where any part of this thesis has previously been submitted for a degree or any other qualification at this University or any other institution, this has been clearly stated.
- Where I have consulted the published work of others, this is always clearly attributed.
- Where I have quoted from the work of others, the source is always given. With the exception of such quotations, this thesis is entirely my own work.
- I have acknowledged all main sources of help.
- Where the thesis is based on work done by myself jointly with others, I have made clear exactly what was done by others and what I have contributed myself.

Signed: Naoyuki Ishida

Date: 2025/12/18

"The unexamined life is not worth living."

Socrates

Acknowledgements

In completing this dissertation, I have received guidance, support, and warm encouragement from countless individuals. I would like to take this opportunity to express my heartfelt gratitude to them all.

First and foremost, I would like to express my deepest gratitude to my supervisor, Professor Kazuhiro Izui. Without his daily enthusiastic guidance and strong support throughout my research activities, it would not have been possible to bring this study to fruition. I am sincerely grateful.

I am indebted to Professor Shinji Nishiwaki in more ways than I can express. Had it not been for my encounter with him, I would not have joined this research region. I am sincerely grateful for his daily research guidance, his detailed care for my personal and mental well-being, and for creating many valuable connections for my future.

I am deeply grateful to Professor Kozo Furuta, who always kindly consulted with me on everything from writing papers to research progress. It was thanks to his precise advice that I was able to overcome the challenges in my research.

I would like to express my sincere gratitude to Dr. Tsuguo Kondoh. Since I joined this laboratory, he has consistently taught me the importance of mathematical rigor. The comments and materials he provided during our numerous meetings have been invaluable to me. I deeply admire his vast knowledge and will continue to strive to reach his level of expertise.

I learned a great deal from Professor Jike Han's approach to research. His example of consistently publishing a high volume of papers was a great source of inspiration for me. I found many aspects of his research methods and philosophy to be incredibly insightful.

I am also very grateful to Professor Masashi Kishimoto and Professor Hiroshi Iwai. They provided me with enthusiastic guidance, even though I was a student from a different laboratory. It is no exaggeration to say that my deep knowledge of batteries is a testament to their guidance. I am sincerely thankful.

I would also like to express my sincere thanks to the senior members, colleagues, and junior members of my laboratory. I am especially grateful to my senior, Dr. Hao Li. When I first joined the lab, communication was difficult due to the COVID-19 pandemic, but Hao patiently taught me the fundamentals of research. The time he spent meticulously reviewing my paper with me until 4 a.m. is a memory I will treasure for the rest of my life. I will never forget the days spent discussing research with Tiannan Fu, Seongwon Bae, Sheng Pan, and Nari Nakayama. Being able to talk with people from diverse backgrounds about topics beyond research has greatly broadened my perspective. Having the reliable Takamitsu Sasaki by my side allowed me to engage in deep discussions about the finer details of my research. Attending the international conference in Lisbon together is one of my fondest memories. I am truly grateful. To Shun Noguchi, although I may have pulled you onto the doctoral path, it was your figure, working diligently and silently, that enabled me to work late into the night. In the process of teaching you various things, I found that there was much for me to learn as well. I burdened you with many miscellaneous tasks, but you accepted them without a single complaint. Thank you.

To my father, mother, grandfathers, and grandmothers: thank you for allowing me to pursue a doctoral degree and for your unwavering financial and mental support. Seeing your happy faces has always been my greatest motivation. I would also like to thank my brother and sister-in-law, Koichi and Riho, and my niece, Anna. Thanks to the three of you living in Kyoto, I was able to live here without feeling lonely. I know I caused a lot of trouble, but thank you for always welcoming me with a smile.

Finally, I would like to express my deepest gratitude to my partner, Kinuko Kubota. I could not have come this far without your unwavering encouragement and support. When I struggled with the lack of results or felt discouraged after a paper rejection, you were always there to lift my spirits. You were genuinely happy when my paper was accepted, perhaps even more than I was. I sincerely hope to walk through the rest of my life together with you.

I am profoundly grateful for the financial support that made this research possible. I would like to extend my sincere appreciation to DMG MORI CO., LTD. for their generous grant. Furthermore, this work was supported by the Japan Society for the Promotion of Science (JSPS) through the Research Fellowship for Young Scientists (DC2). This invaluable assistance provided me with a stable environment, allowing me to devote myself fully to my research without financial concern. I wish to express my deepest thanks to these organizations and to all the taxpayers whose contributions fund these vital programs.

1. Scholarship from DMG MORI CO., LTD (4/1/2023–3/31/2025)
2. Grant-in-Aid for the Japan Society for the Promotion of Science Research Fellow (4/1/2025–)

Naoyuki Ishida
Kyoto, December, 2025

KYOTO UNIVERSITY

Abstract

Strength of Materials and Structures Lab
Graduate School of Engineering

Doctor of Engineering

An optimum design methodology for battery component structures using topology optimization

by Naoyuki Ishida

The growing demand for high-performance batteries challenges traditional development methods. This dissertation establishes a physics-based design framework using topology optimization (TO), proposing it as a generative tool to discover non-intuitive architectures and accelerate research and development. The research addresses linking multi-scale material distributions to macro-scale properties and formulating efficient sensitivity analysis for coupled multiphysics problems, applying the methodologies to three distinct design cases. First, a TO method for a lithium-ion battery separator successfully suppressed dendrite-induced short circuits by maximizing ion path tortuosity. Second, for all-solid-state batteries (ASSBs), the framework generated novel, layered microstructures by coupling homogenization with a time-dependent model, using a new objective function that reduced computational cost. Third, a meso-scale optimization method for an ASSB anode, using a novel data-driven material interpolation, yielded functionally graded electrodes that significantly enhanced performance, reducing overvoltage by 12.4% and increasing capacity by 6.81%. In summary, this dissertation pioneers the application of TO to complex, multi-scale design problems in batteries. By transitioning from simulation-based analysis to physics-based generative design, this work provides a robust and adaptable computational framework. This framework, which integrates multi-physics models with novel data-driven approaches, enables the discovery of non-intuitive, high-performance architectures. It thereby contributes a vital tool to accelerate the development of next-generation energy storage systems tailored to meet diverse and demanding application requirements.

Keywords: topology optimization, battery separator, all-solid-state-battery, adjoint sensitivity analysis, strongly-coupled problem

Contents

Acknowledgements	vii
Abstract	ix
List of Figures	xv
List of Tables	xix
1 General Introduction	1
1.1 Introduction	1
1.1.1 General background	1
1.1.2 Motivation	2
1.1.3 Problem Statement and Research Objectives	4
1.1.4 Thesis structure	6
1.2 Topology optimization	7
1.2.1 Structure representation method	7
Binary representation	7
Density method	9
Level-set method	9
1.2.2 Sensitivity analysis	10
Finite difference method	10
Direct difference method	11
Adjoint method	12
1.2.3 Update scheme	13
The Method of Moving Asymptotes	14
Reaction-diffusion equation	16
Hamilton-Jacobi equation	17
1.2.4 Summary	17
1.3 Structural optimization method for the design of batteries	18
1.3.1 Thermal management	18
1.3.2 Flow battery	19
1.3.3 Solid Oxide Fuel Cell (SOFC)	20
1.3.4 Lithium ion battery	21
1.3.5 Summary	23

2	Topology optimization for the design of battery separators	25
2.1	Introduction	25
2.2	Formulation	27
2.2.1	Design variable	27
2.2.2	Governing equations	30
2.2.3	Optimization problem	31
2.2.4	Sensitivity analysis	34
2.3	Implementation details	35
2.3.1	Optimization algorithm	35
2.4	Numerical examples	36
2.4.1	Verification of sensitivity derivation	37
2.4.2	Optimized configurations in 2D	38
2.4.3	Optimized configurations in 3D	44
2.4.4	Investigation for the objective function	47
2.5	Conclusions	49
3	Topology optimization for all-solid-state-batteries using homogenization method	51
3.1	Introduction	51
3.2	Governing equations	54
3.2.1	Analysis for all-solid-state-batteries	54
3.2.2	Homogenization method	57
3.3	Topology optimization	59
3.3.1	Design variable settings	59
3.3.2	Formulation of optimization problem	59
3.3.3	Update scheme of design variable	63
3.3.4	Optimization algorithm	63
3.4	Numerical examples	64
3.4.1	Problem settings	65
3.4.2	Validation of proposed objective function	66
3.4.3	Verification of derived sensitivity	69
3.4.4	Numerical results	71
	Various C rates	71
	Effect on initial configurations	72
	Effect on macrostructure configurations	74
	Considering two types of unit cells	75
3.5	Conclusions	77
4	Data-driven TO of ASSBs considering conductive additive material	79
4.1	Introduction	80
4.2	Formulation	84
4.2.1	Design variables settings	84

4.2.2	Governing equations	85
4.2.3	Optimization problem	89
4.3	Material interpolation scheme	92
4.3.1	Random material distribution schemes	92
4.3.2	Estimation of normalized transport coefficient and surface area	95
4.3.3	Algorithm for obtaining interpolation functions	98
4.4	Numerical implementations	101
4.5	Numerical examples	102
4.5.1	Numerical settings	102
4.5.2	Material interpolation function profile	102
4.5.3	Optimization results	110
	Performance evaluation of optimized material distribution: Comparison with uniform distribution	111
	Comparison and analysis of optimal solutions obtained using different ma- terial interpolation functions	114
	Relation between electronic conductivity values of materials and optimized structures	116
4.6	Conclusions	116
5	General Conclusions	119
5.1	Summary of the Dissertation	119
5.2	Overall Significance and Implications	121
5.3	Limitations and Future Research Directions	122
5.3.1	Limitations of the Current Work	122
5.3.2	Recommendations for Future Research	122
A	Sensitivity Analysis	125
A.1	Minimal heat compliance problem with Helmholtz-type filtering	125
A.2	Sensitivity analysis for ASSBs problem	129
B	Implementation details	133
B.1	Random material distribution schemes and estimation of normalized transport co- efficient	133
B.2	Least square with constraint	146
	References	155
	About the author	165
	Publication List	167

List of Figures

1.1	Structure representation methods	8
2.1	The configurations of a battery	28
2.2	The definition of the unit cell	28
2.3	Definition of the plane A	32
2.4	Schematics of dendrite growth in two-dimensions	32
2.5	Optimization flow chart.	36
2.6	The definition of the unit cells and discretization	37
2.7	The comparison of the discrete sensitivity distribution $S_{f,\text{auto}}$ and the distribution of the product of the continuous sensitivity and the element area $S_f \times h^2$ for three types of design variable distributions	39
2.8	The configurations of the initial and optimized design variables, and the distributions of the concentration and flux (black arrow) for #1, #2, and #3	40
2.9	Objective function histories for #1 to #3	41
2.10	The analysis domain for evaluating the tortuosity factor of optimized separator configurations (a), and symmetrically aligned optimal structures with only the electrolyte region extracted (b–d)	42
2.11	The optimized separator configurations, the ion flux, and the concentration distributions for #4, #5, and #6	45
2.12	Comparison of computational costs with varying numbers of discretized elements in condition #4	47
2.13	Optimized design variable configurations and distributions of concentration and flux (black arrows) for optimization problems with objective functions J and J_t	48
3.1	Schematics of the all-solid-state-battery	55
3.2	Schematics of the homogenization method	57
3.3	Design variable in the unit cell	59
3.4	Charge-discharge curves	60
3.5	Optimization flow chart.	64
3.6	Schematics of the macro scale structure, type B	65
3.7	Meshes for each scale geometry	66
3.8	Plots for the normalized proposed and original objective functions for 0.5C, 1C and 2C across varying macrostructure configurations with Q [8, 10, 12, 14] and n [1, 2, 3, 4, 5]	68

3.9	Material distributions within the unit cell when the gradient of the distributed active material is 0.5	69
3.10	Plots for the normalized proposed and original objective functions for 1C when the angle θ between the y_1 -axis and the active material distribution in the unit cell is changed	69
3.11	Design variable distribution ρ and the cut lines for the comparison of the derived sensitivity and the numerical difference	70
3.12	Plots for the normalized sensitivity J' and numerical difference $\tilde{J}_{pr}$ along the cut lines	70
3.13	The obtained results for 0.5C, 1C and 2C	71
3.14	The initial distributions in the unit cell for the optimization	73
3.15	The obtained results for each initial distribution	73
3.16	Plots for the objective and constraint function histories for the initial distributions .	74
3.17	The obtained results for each macro scale structure	75
3.18	Schematic of two unit cells considered in the composite anode	75
3.19	The obtained results for two unit cells, Y_1 and Y_2 , considered in the composite anode	76
3.20	Concentration distributions upon termination of the calculation for the discharging process when considering one and two types of unit cells	76
4.1	Model of all-solid-state-batteries	84
4.2	Domains and boundaries definitions	85
4.3	Model of the microstructure analysis	93
4.4	Randomly generated material distributions for single type of material in a unit cell with R_s of 0.2 and a standard deviation s of 0.10	94
4.5	Randomly generated material distributions in a unit cell for two material types: $R_{s1} = 0.1$ (red) and $R_{s2} = 0.2$ (gray) with a standard deviation $s = 0.10$	96
4.6	Algorithm for making the material interpolation function H_1 and A	98
4.7	Algorithm for the optimization	101
4.8	Mesh for the unit cell	104
4.9	Histograms of the sampled 100 points and probability density functions $f(R_s, s)$ of log-normal distributions	105
4.10	Comparison of data-points from microstructure analysis and the approximated material interpolation functions, $\tilde{H}_1$, $\tilde{H}_2$ and $\tilde{A}$ with $R_s = 0.2$ and $s = 0.1$	106
4.11	Visualization of absolute (vertical axis) and relative (color) errors between data points and approximated material interpolation function	107
4.12	Plots of the approximated material interpolation function H_1 for the dimensionless transport coefficient with $R_s = 0.2, 0.1$, and 0.04 and $s = 0.1$, and SIMP method with $p = 1.5, 2$, and 3 as a function of volume fraction	109
4.13	Plots of the obtained data points with $R_s = 0.04$ and $s = 0.1$, and the function profile corresponding to $p_t = 0.5153$ in Eq. 4.105	110
4.14	Mesh for the battery analysis	110
4.15	The initial material distributions for the optimization	111

4.16	The obtained material distributions by the proposed method	111
4.17	History plots of objective and constraint values during the optimization iterations .	112
4.18	The uniform material configurations of rectangular-type and parallel-type for the comparison	113
4.19	Histories of $U_p(\tilde{c}_p)$ and discharge curve $\tilde{\phi}_s$ during the discharge process for the uniform rectangular-type, uniform parallel-type and optimized material distributions as functions of SOC_n	113
4.20	The obtained material distributions by the optimization method using the proposed function H_1 and H_2 , and the penalty parameter p of 3 and 1.5 with SIMP method . .	115
4.21	The obtained material distributions for the various electronic conductivity values of the active material and conductive additive material: $(\sigma_n, \sigma_c)=(0.001$ [S/m], 1 [S/m]), (0.01 [S/m], 10 [S/m]), (0.1 [S/m], 100 [S/m]), and (1 [S/m], 1000 [S/m]) . .	118

List of Tables

2.1	The volume fraction of electrolyte regions within the unit cells, and tortuosity factor of the optimized separator structures obtained from the condition, #1, #2, and #3 . . .	44
2.2	The set constraint values and the final objective function values for each condition, from #4 to #6	44
3.1	Boundary conditions	56
3.2	Parameter settings	67
3.3	Original objective function values J_{ori} analyzed at 0.5C, 1C, and 2C for structures obtained at 0.5C, 1C, and 2C in the optimization, respectively	72
4.1	List of variables used in the paper	83
4.2	Parameter settings	103
4.3	The coefficients h for $\tilde{H}_1$ obtained by least squares for different values U and the values of AIC	107
4.4	The coefficients w for $\tilde{A}$ obtained by least squares for different values U and the values of AIC	107
4.5	AIC values for varying orders of least squares, U_a and U_c	107

Dedicated to my parents and my family
To my friends

Chapter 1

General Introduction

CONTENTS

1.1 Introduction	1	Adjoint method	12
1.1.1 General background	1	1.2.3 Update scheme	13
1.1.2 Motivation	2	The Method of Moving Asymptotes	14
1.1.3 Problem Statement and Research Objectives	4	Reaction-diffusion equation	16
1.1.4 Thesis structure	6	Hamilton-Jacobi equation	17
1.2 Topology optimization	7	1.2.4 Summary	17
1.2.1 Structure representation method	7	1.3 Structural optimization method for the design of batteries	18
Binary representation	7	1.3.1 Thermal management	18
Density method	9	1.3.2 Flow battery	19
Level-set method	9	1.3.3 Solid Oxide Fuel Cell (SOFC)	20
1.2.2 Sensitivity analysis	10	1.3.4 Lithium ion battery	21
Finite difference method	10	1.3.5 Summary	23
Direct difference method	11		

1.1 Introduction

1.1.1 General background

Addressing climate change, one of the most pressing challenges facing modern society, necessitates a fundamental transformation of the global energy system towards decarbonization. In achieving this transformation, electrochemical energy storage technologies, particularly high performance secondary batteries, are regarded not merely as incremental improvements but as a keystone technology enabling multiple global megatrends simultaneously.

First, in the energy sector, the adoption of renewable energy sources such as solar and wind is rapidly expanding to reduce CO₂ emissions. However, the intermittent nature of these sources, which are dependent on weather conditions, can compromise the stability of the power grid (Wu et al., 2022; Gowrisankaran et al., 2016). Overcoming this challenge and maintaining a stable electricity supply requires large-scale energy storage systems to balance fluctuations between generation and demand. Recent technological innovations have dramatically improved the economics of energy storage systems. Notably, the costs of solar photovoltaics, wind power, and lithium-ion batteries have fallen drastically. For instance, the levelized cost of energy for systems combining solar PV and lithium-ion batteries reportedly decreased by 89% between 2010 and 2021 (Lazard, 2021). This cost reduction signifies that the combination of renewable energy and battery storage has become directly cost-competitive with new fossil fuel power plants (Schmidt and Staffell, 2023). Furthermore, the role of battery storage extends beyond merely replacing new power plants. It also shows the potential to replace natural gas-fired "peaker plants," which operate only during times of highest electricity demand, at a lower cost. One study estimates that under specific conditions (up to a 17% capacity factor), a peaker system using hydrogen fuel cells could be more cost-effective than conventional fossil-fuel peaker plants (Chojkiewicz and Phadke, 2024). While economic challenges remain in completely replacing all high-demand scenarios with batteries, it is evident that energy storage technology is fundamentally altering the economics of the power system. These facts demonstrate that energy storage is not just a technical solution but a powerful driver of structural transformation in the energy economy.

Second, the transportation sector is undergoing rapid electrification to move away from its dependence on fossil fuels. Electric vehicles (EVs) are at the forefront of this transformation, and their market penetration is directly dependent on advancements in battery technology that deliver longer driving ranges, faster charging times, and lower costs. Driven by EV passenger cars, light-duty trucks, and heavy-duty trucks, the demand for batteries is projected to grow exponentially (Kim et al., 2022). In addition to these large-scale applications, the proliferation of portable electronics, such as smartphones and wearable devices, continues to drive demand for smaller, safer batteries with higher energy densities (Antony Jose et al., 2025).

1.1.2 Motivation

The convergence of demand from these multiple large sectors is inducing unprecedented investment in the battery ecosystem and accelerating research and development (Ziegler and Trancik, 2021). This has led to significant progress towards the commercialization of not only existing lithium-ion batteries (LIBs) but also next-generation technologies such as all-solid-state batteries (ASSBs) (Sung et al., 2024). However, this expansion in demand brings not only an increase in volume but also a diversification in quality. For example, the performance requirements for a battery optimized for plant storage (prioritizing cost and cycle life) differ fundamentally from those for a battery designed for electric vehicle (prioritizing specific power and safety) or for a smartphone

(prioritizing volumetric energy density). This suggests the limitations of a generic development approach and implies that a diverse portfolio of application-specific batteries is essential. This increasing complexity of design requirements highlights the limitations of traditional development methods, which necessitate vast and time-consuming empirical optimization cycles for each new application or material. Therefore, the establishment of a systematic and adaptive design methodology to meet these diversifying demands swiftly and efficiently is urgently required.

Numerical simulation is a powerful tool for tackling this challenge. Mathematically modeling the behavior of a battery is crucial for predicting and estimating its real-time operation, which in turn contributes to improving charging/discharging technologies and understanding degradation mechanisms. A landmark achievement in this field is the electrochemical model proposed by Doyle, Fuller, and Newman, commonly known as the "DFN model" or "Pseudo-Two-Dimensional (P2D) model" (Doyle et al., 1993). The DFN model describes the intercalation of lithium into active material particles within the electrode (microscale) and ion transportation in the electrolyte (macroscale) using a set of partial differential and algebraic equations. This model, despite its complexity involving over 30 physical and chemical parameters, can reproduce the internal physical phenomena of a battery with high fidelity, making it widely used as a benchmark.

Underlying approaches such as the DFN model is the porous electrode theory pioneered by John Newman and his colleagues (Newman and Thomas-Alyea, 2004). This theory does not directly compute the complex microstructure composed of active material particles and pores. Instead, it employs a volume-averaging (homogenization) technique. This allows the electrode to be treated as a homogeneous continuum where multiple phases with different properties (e.g., active material phase and electrolyte phase) overlap. Through this homogenization approach, macroscopic transport phenomena of ions and electrons across the entire electrode can be described using averaged physical properties such as porosity and tortuosity, making cell-level performance analysis computationally feasible.

To further advance these analytical techniques and to more actively contribute to battery performance enhancement, this research focuses on structural optimization, particularly topology optimization (TO). Structural optimization is a collective term for computational methods that find the optimal shape or material layout to maximize the performance of a structure under given constraints (Bendsøe and Sigmund, 2004). It is generally classified into three categories: "size optimization," where design variables are cross-sectional areas or thicknesses; "shape optimization," which modifies the boundary shape of a structure; and "topology optimization," which alters the boundary shape and the number of holes to find the optimal material layout from scratch. Among these, TO offers the highest degree of design freedom as it does not depend on an initial shape, holding the potential to create innovative structures that often surpass human intuition.

The theoretical foundations of TO can be traced back to the late 19th and early 20th centuries, with studies by Maxwell and Michell on the optimal design of truss structures Michell, 1904. However, its development into a practical design tool had to await advances in computational

science. The dawn of modern TO was marked by a groundbreaking paper in 1988 by Bendsøe and Kikuchi, which used a homogenization method Bendsøe and Kikuchi, 1988. They proposed a new formulation by considering the design domain as a composite material with innumerable microscopic voids and continuously varying its material density from 0 to 1 to find the optimal material distribution. This research was revolutionary in that it formulated TO, previously treated as a discrete 0-1 problem, as a continuum problem. It spurred the development of various subsequent methods such as the Solid Isotropic Material with Penalization (SIMP) method (Bendsøe and Sigmund, 1999) and the level-set method (Wang et al., 2003), attracting significant attention from both academia and industry. As a result, TO has expanded its application beyond its original field of structural mechanics to a diverse range of physical problems.

For example, in fluid dynamics, it has been applied to the design of channels that minimize pressure drop (Borrvall and Petersson, 2003). In thermal engineering, it is used for designing heat sinks and heat exchangers that maximize heat transfer efficiency (Gersborg-Hansen et al., 2006; Matsumori et al., 2013). Furthermore, in the fields of electromagnetics and photonics, its utility is widely recognized for applications such as the design of photonic band-gap materials that block or transmit electromagnetic waves in specific frequency bands (Sigmund and Søndergaard Jensen, 2003), and for enhancing the performance of optical devices like nano-antennas and waveguides (Jensen and Sigmund, 2011).

1.1.3 Problem Statement and Research Objectives

Despite its great success, the application of topology optimization has not yet been sufficiently extended to the domain of internal structural design of lithium-ion batteries.

Many research achievements concerning various types of batteries have been made using TO. Iwai et al., 2011 proposed a TO method aimed at improving the efficiency of Solid Oxide Fuel Cells (SOFC), demonstrating its potential in applications. Yaji et al., 2018, Lin et al., 2022 and Roy et al., 2022 dealt with redox flow batteries, which have liquid electrolytes for the anode and cathode. They proposed a TO method to optimize the flow path inside redox batteries, further improving their efficiency. A TO method has also been proposed for the cooling plate to manage heat, which is problematic in batteries (Mo et al., 2021; Guo et al., 2022a). Pejman and Najafi have proposed the TO method for the structural battery composite (SBC) by assuming physics as the equivalent network (Pejman and Najafi, 2023). The SBC represents an innovative category of multifunctional materials, merging carbon fiber composites' load-supporting properties with a lithium-ion battery's energy storage function (Snyder et al., 2007; Snyder et al., 2008; Wong et al., 2007; Liu et al., 2009). Moreover, Alizadeh et al., 2023 have proposed the TO method for a porous reactor, which focused on the distribution of the volume fraction of the active and electrolyte materials. More detailed review for TO methods for the design of batteries is provided in Section 1.3. Although the

above-mentioned previous studies have achieved a great deal, they did not derive the accompanying sensitivity in the continuous system in the electrochemical model or examine the transport parameters themselves.

Therefore, this dissertation aims to establish a topology optimization method for the internal components of batteries and to elucidate the mechanisms of performance improvement. To achieve this goal, there are two main challenges to overcome.

The first challenge is the modeling of effective material properties, particularly the appropriate handling of the effective diffusion coefficient, i.e., the tortuosity of the electrode. In topology optimization, the presence or absence (or density) of material at each point in the design domain is a design variable. Consequently, a physically sound constitutive law that links this microscale material distribution to the macroscale effective diffusion coefficient governing ion transport is indispensable. Establishing this relationship purely through experiments is not feasible given the vast design space of electrode structures. To address this problem, this study employs computational science techniques, such as the homogenization method (Allaire, 1992), a mathematical theory that connects microstructure to equivalent properties, and data-driven approaches based on numerical experiments. The homogenization method is a mathematical technique for deriving the equivalent (homogenized) physical properties of a composite material with a microscopically periodic structure, thereby enabling a theoretical description of the relationship between material density and effective diffusion coefficient.

The second challenge is the efficient sensitivity analysis for complex coupled problems. A battery system is a typical multiphysics problem where concentration, potential, and temperature fields interact with each other. Furthermore, since it deals with the charge-discharge processes of a secondary battery, it is essential to consider time-dependency, i.e., unsteadiness. To perform gradient-based optimization for such a complex problem, it is necessary to efficiently calculate the "sensitivity," which indicates how the objective function changes with respect to a vast number of design variables. In this sensitivity derivation, the continuous adjoint sensitivity analysis is an extremely powerful method that can calculate all sensitivities in a single analysis, independent of the number of design variables (Giles and Pierce, 2000). Particularly for unsteady problems, by appropriately formulating the adjoint problem, it is possible to derive the sensitivity without storing information from all time steps of the forward calculation, thus avoiding memory consumption issues. Therefore, in this dissertation, we derive the adjoint equations in a continuous system for all proposed optimization methods to achieve efficient gradient computation.

1.1.4 Thesis structure

Based on the background and problem awareness described above, this dissertation is structured as follows. The remainder of this section provides an overview of topology optimization in Section 1.2 and structural optimization for the design of batteries in Section 1.3.

Chapter 2 establishes a topology optimization method for the internal structure of a lithium-ion battery separator. The separator is a thin, porous membrane component soaked in electrolyte and positioned between the electrodes. It serves multiple, often conflicting, roles: ensuring an ion transport path while preventing electrical short circuits caused by dendrite growth or electrode contact. To maximize its performance, this chapter defines a unit cell and optimizes its internal structure. The formulation aims to suppress linear dendrite growth between electrodes by creating a tortuous ion transport path. This is achieved by maximizing the magnitude of the in-plane ion concentration gradient, which is perpendicular to the through-plane direction. At this stage, the macroscopic behavior of the entire battery is not considered; the focus is solely on the effective diffusion coefficient within the unit cell microstructure to derive its optimal structure.

Chapter 3 develops a topology optimization method for the microstructure of composite electrodes in all-solid-state batteries (ASSBs). ASSBs are anticipated as a next-generation technology that can bring about dramatic improvements in safety and energy density by replacing flammable liquid electrolytes with non-flammable solid electrolytes. However, a challenge arises because both the active material and the solid electrolyte are solids, making it difficult to secure sufficient contact area between them, which leads to a decrease in ionic and electronic conductivity. To solve this problem, a configuration is widely adopted where a composite region, a mixture of active material and solid electrolyte, is placed at the interface between the electrode and the solid electrolyte. This chapter, therefore, designs the optimal material mixing state, i.e., the microscopic material distribution, within this composite electrode. Here, the homogenization method is introduced as a technique to link the physical properties of the microstructure to the macroscopic effective properties. That is, we assume that the composite region is composed of a collection of periodic unit cells and seek the optimal material distribution within that unit cell. Unlike Chapter 2, this chapter also solves the macroscale equations considering the nonlinear coupled problem of concentration and potential fields to evaluate the performance of the entire electrode and performs time-dependent optimization. The derivative of the objective function with respect to the design variables is analytically derived based on the adjoint variable method.

Chapter 4 focuses on meso-scale optimization, differing from the approach in Chapter 3. The method proposed in Chapter 3 directly designs the detailed microstructure within a unit cell, which poses a challenge for realization with current manufacturing technologies. Therefore, to obtain a more practical design, this chapter treats the volume fractions of each material within the composite electrode as design variables. In other words, instead of focusing on the detailed

structure within a unit cell, we focus on the amount of material at each point and derive its optimal spatial distribution. Crucial to establishing this approach is the construction of a material interpolation function that relates the volume fractions to the effective transport coefficients. The homogenization method used in Chapter 3 assumes a determined material distribution within the unit cell and is not applicable to the approach in this chapter. Therefore, this study proposes a new method for constructing a physically-based interpolation function in a data-driven manner. Specifically, we first generate a large number of microstructures with randomly placed materials in a virtual unit cell on a computer and calculate their respective effective transport coefficients through numerical analysis. Then, using the obtained large dataset (pairs of volume fraction and effective transport coefficient), an interpolation function is created using a least-squares method. The novelty of this method lies in imposing positivity and monotonic increase constraints on the function to ensure the stability of the optimization calculation and to obtain a physically meaningful solution.

Finally, Chapter 5 summarizes the findings of this research and discusses future prospects.

1.2 Topology optimization

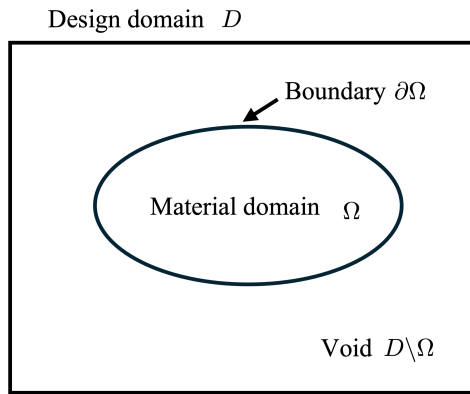
This section provides an overview of topology optimization. Topology optimization is a method that reformulates a structural design problem as a material distribution problem. Section 1.2.1 describes the material representation methods currently in use. Then, Section 1.2.2 summarizes the methods for deriving sensitivities, which are the derivatives of the objective function with respect to the design variables, necessary for finding better shapes. Finally, Section 1.2.3 classifies the methods for updating the design variables defined in the previous section.

1.2.1 Structure representation method

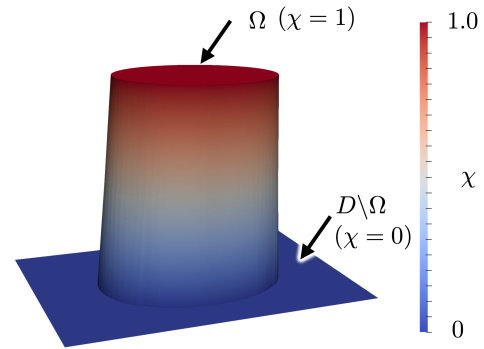
Binary representation

The most intuitive method is to use a characteristic function $\chi(x)$. As shown in the following equation, this method assigns a value of 1 to each point x in the design domain D if it belongs to the material domain Ω , and 0 if it belongs to the void domain $D \setminus \Omega$ as shown in Fig. 1.1.

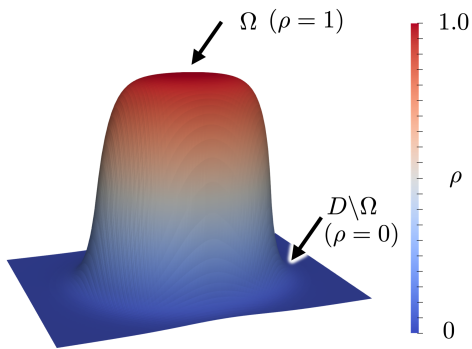
$$\chi(x) = \begin{cases} 1 & \text{if } x \in \Omega \\ 0 & \text{if } x \in D \setminus \Omega. \end{cases} \quad (1.1)$$



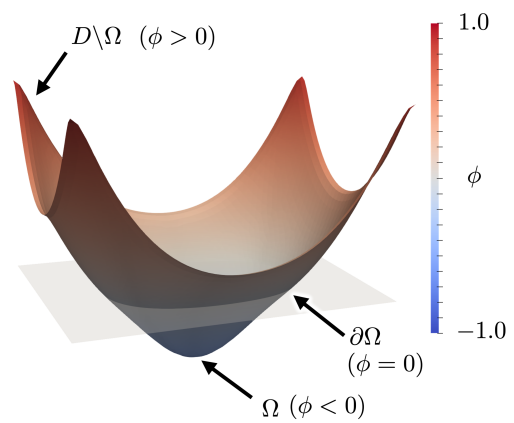
(A) Schematics of the computational domain



(B) Binary representation



(C) Density approach



(D) For micro scale

FIGURE 1.1: Structure representation methods

Although this is the most fundamental concept, it can lead to discontinuous optimal structures where 0 and 1 are distributed alternately within the domain, as has been shown in previous studies.

Density method

The density approach is a continuous extension of the characteristic function, where the density ρ can take any value between 0 and 1 ($\rho \in [0 : 1]$). That is, $\rho = 1$ in the material domain, $\rho = 0$ in the void domain, and the density function takes values between 0 and 1 in the intermediate regions as shown in Fig. 1.1c. Using this density function, physical properties such as the diffusion coefficient or Young's modulus are interpolated with a function as shown in the following equation.

$$K = (K_1 - K_0)\rho^p + K_0. \quad (1.2)$$

This method is often called the Solid Isotropic Material with Penalization (SIMP) method. It interpolates the physical property to be K_1 where material exists and K_0 where it does not. Assigning a small physical property to the void region is done to stabilize the numerical analysis. The density approach is the most commonly used method due to its simplicity in implementation and formulation. However, a problem with this method is the difficulty in interpreting the intermediate material, known as "grayscale."

Level-set method

The level-set method is a way to eliminate grayscale while maintaining continuity by explicitly defining the boundary between material and void with a function. In the level-set method, we consider a level-set function $\phi(x)$ that is negative (or positive) in the material domain, positive (or negative) in the void domain, and zero at the boundary, as shown in the following equation and Fig. 1.1d:

$$\begin{cases} \phi(x) < 0 & \text{if } x \in \Omega \\ \phi(x) = 0 & \text{if } x \in \partial\Omega \\ \phi(x) > 0 & \text{if } x \in D \setminus \Omega. \end{cases} \quad (1.3)$$

In this method, since the boundary is explicitly defined in the resulting optimal distribution, it facilitates the generation of CAD models in the post-processing stage. On the other hand, because the material is sharply divided during the optimization process, numerical calculations often become unstable, frequently requiring additional methods such as mesh adaptation or XFEM[Haosan paper].

1.2.2 Sensitivity analysis

TO does not directly yield the optimal solution in a single step; instead, it iteratively updates the design variables formulated in the previous section to obtain an optimal solution—more precisely, a local optimum. The TO methods are generally classified into two categories: gradient-based methods and non-gradient-based methods. In gradient-based methods, the sensitivities, i.e., the derivatives of the objective function with respect to the design variables, are computed, and the design variables are iteratively updated based on this gradient information to search the optimum. In contrast, non-gradient-based methods do not rely on sensitivities but instead employ heuristic algorithms, such as genetic algorithms or differential evolution, to update the design variables. Gradient-based methods are known to be computationally more efficient and cost-effective than their non-gradient-based counterparts. On the other hand, non-gradient-based methods are capable of exploring global optima and, during the optimization process, can provide multiple local optima simultaneously. This characteristic is particularly advantageous in the conceptual design stage, where obtaining diverse design guidelines within a single optimization run is valuable. In this section, the focus is placed on gradient-based methods, which have played a central role in the advancement of TO. Specifically, three representative approaches for deriving sensitivities, which form the foundation of these methods, are introduced and discussed.

For the sake of explanation, consider the following optimization problem in the discretized form:

$$\begin{aligned} \min_{\mathbf{x} \in \chi} \quad & F(\mathbf{x}, \mathbf{u}(\mathbf{x})) \quad \mathbf{x} = [x_1, x_2, \dots, x_n]^T \\ \text{s.t.} \quad & \mathbf{K}\mathbf{u} = \mathbf{f}, \end{aligned} \tag{1.4}$$

where $\mathbf{x}$ denotes the design variable defined at each discretized node, representing the structural configuration, and $\mathbf{u}$ denotes the displacement field governed by the constraint equation. $\mathbf{K}$ is the globally assembled stiffness matrix, and $\mathbf{f}$ is the external force vector.

Finite difference method

One of the most fundamental approaches for sensitivity analysis is the finite difference method. By applying a Taylor series expansion, the objective function can be approximated as follows:

$$\begin{aligned} F(\mathbf{x} + \varepsilon_i) &= F(\mathbf{x}) + \frac{dF}{dx_i} \varepsilon_i + \frac{1}{2} \frac{d^2F}{dx_i^2} \varepsilon_i^2 + O(\varepsilon^3) \\ F(\mathbf{x} - \varepsilon_i) &= F(\mathbf{x}) - \frac{dF}{dx_i} \varepsilon_i + \frac{1}{2} \frac{d^2F}{dx_i^2} \varepsilon_i^2 + O(\varepsilon^3). \end{aligned} \tag{1.5}$$

Here, ε_i represents a perturbation applied to the i -th node. Then, we can approximate the derivative as follows:

$$\frac{dF}{dx_i} = \frac{F(\mathbf{x} + \varepsilon_i) - F(\mathbf{x} - \varepsilon_i)}{2\varepsilon_i}. \quad (1.6)$$

Accordingly, Eq. (1.6) expresses the sensitivity of the objective function with respect to the i -th design variable. The finite difference method has the advantage that sensitivities can be derived without requiring explicit differentiation, relying instead solely on repeated state analyses, which makes the derivation relatively straightforward.

Several variants of the finite difference scheme exist: forward difference, where the design variable is perturbed in the positive direction; backward difference, where it is perturbed in the negative direction; and central difference, where perturbations are applied in both directions to achieve higher accuracy. Among these, the central difference scheme generally provides improved accuracy and reduces the effect of round-off errors, albeit at the cost of additional computational effort compared to forward or backward difference.

Nevertheless, in all cases, ε must be chosen sufficiently small to approximate the derivative accurately. If ε is excessively small, numerical round-off errors may dominate, leading to inaccurate sensitivities. Moreover, since the number of required analyses scales proportionally with twice the number of design variables, the computational cost becomes prohibitively high. For this reason, the finite difference method is rarely adopted in large-scale topology optimization.

Direct difference method

Next, the direct differentiation method is introduced. In general, the objective function depends on both the design variables and state variables such as the temperature or displacement fields. Since these state variables are themselves dependent on the structural configuration, they also depend on the design variables. Taking this dependency into account, the derivative of the objective function with respect to the design variables can be expressed using the chain rule as follows:

$$\frac{dF}{dx} = \frac{\partial F}{\partial x} + \frac{\partial F}{\partial \mathbf{u}} \frac{\partial \mathbf{u}}{\partial x}. \quad (1.7)$$

Here, directly evaluating $\frac{\partial \mathbf{u}}{\partial x}$ is generally difficult. To address this, attention is directed to the governing equation that defines $\mathbf{u}$. Differentiating the governing equation with respect to the design variable yields the following relation:

$$\frac{\partial \mathbf{K}}{\partial x_i} \mathbf{u} + \mathbf{K} \frac{\partial \mathbf{u}}{\partial x_i} = \frac{\partial \mathbf{f}}{\partial x_i}. \quad (1.8)$$

where x_i denotes the i -th design variable. This equation can be further rearranged as:

$$\begin{aligned} K \frac{\partial u}{\partial x_i} &= -\frac{\partial K}{\partial x_i} u + \frac{\partial f}{\partial x_i} \\ \Rightarrow \frac{\partial u}{\partial x_i} &= K^{-1} \left(-\frac{\partial K}{\partial x_i} u + \frac{\partial f}{\partial x_i} \right). \end{aligned} \quad (1.9)$$

Since the right-hand side of Eq. (1.9) can be derived analytically, $\frac{\partial u}{\partial x}$ can be obtained accordingly. Substituting this term into Eq. (1.7) then enables the evaluation of sensitivities.

However, the drawback of this approach is that the computation corresponding to Eq. (1.9) must be performed for each design variable. As a result, the computational cost increases linearly with the number of design variables, making this method impractical for large-scale topology optimization problems, where the number of design variables is typically very large.

Adjoint method

Finally, the adjoint method is introduced. This method was developed to overcome the high computational cost inherent in the finite difference method and the direct differentiation method discussed previously. By introducing adjoint variables, the adjoint method allows for sensitivity analysis in a form independent of the number of design variables, thereby enabling efficient evaluations even in large-scale topology optimization problems.

The fundamental concept of the adjoint method is to introduce an adjoint variable λ and define an extended Lagrangian for the objective function:

$$\mathcal{L} = F + \lambda^T (K\mathbf{u} - \mathbf{f}). \quad (1.10)$$

The sensitivity can then be derived from the Lagrangian as:

$$\frac{dF}{dx} \equiv \frac{d\mathcal{L}}{dx} = \frac{\partial \mathcal{L}}{\partial x} + \frac{\partial \mathcal{L}}{\partial \mathbf{u}} \frac{\partial \mathbf{u}}{\partial x}. \quad (1.11)$$

As noted in the direct differentiation method, directly evaluating the matrix $\frac{\partial \mathbf{u}}{\partial x}$ is generally intractable. While the direct differentiation method circumvents this difficulty by differentiating the governing equation with respect to the design variables, the adjoint method eliminates the need to consider this term by appropriately defining the adjoint variable such that the associated coefficient $\frac{\partial \mathcal{L}}{\partial \mathbf{u}}$ vanishes, as shown in Eq. (1.11). The expression for $\frac{\partial \mathcal{L}}{\partial \mathbf{u}}$ is given by:

$$\begin{aligned} \frac{\partial \mathcal{L}}{\partial \mathbf{u}} &= K^T \lambda - \frac{\partial F}{\partial \mathbf{u}} = 0 \\ \Rightarrow K^T \lambda &= \frac{\partial F}{\partial \mathbf{u}}. \end{aligned} \quad (1.12)$$

Thus, by solving the adjoint equation, Eq. (1.12), governing the adjoint variable λ and substituting the result into the sensitivity expression, only the first term on the right-hand side of Eq. (1.11) needs to be evaluated. Notably, when the coefficient $\frac{\partial F}{\partial u}$ corresponds to the external force vector f , the adjoint equation coincides with the governing equation. In this case, the adjoint and state variables become identical, and no additional adjoint computation is required. Such problems are referred to as self-adjoint problems, and formulating the objective function in this way can significantly reduce the computational cost.

There are two major approaches for deriving sensitivities using the adjoint method. The first is based on automatic differentiation (AD). In this approach, all dependencies and intermediate values from the evaluation of the governing equation and the objective function are stored, and derivatives are computed using backpropagation. A key advantage of AD is that the sensitivity derivation does not need to be reformulated when the objective function changes, making it highly versatile and suitable for multi-physics applications. For this reason, AD-based adjoint methods are widely employed in optimization frameworks and are implemented in commercial software such as COMSOL. However, the drawback is the excessive memory consumption due to the need to store the entire computational history, which poses challenges for large-scale problems.

The second approach is the continuous adjoint method. Here, the adjoint formulation is applied at the continuous level prior to discretization. The adjoint equation derived in this manner is then discretized and solved, independently of the discretization scheme of the governing equation. This independence avoids the need to store all intermediate computations, thereby reducing memory requirements. On the other hand, the sensitivity derivation must be repeated whenever the objective function or governing equations are reformulated, which is a limitation of this approach.

All of the methods proposed in this dissertation are based on continuous adjoint sensitivity analysis.

1.2.3 Update scheme

Next, we describe the methods for updating the design variables. As mentioned above, TO method is an iterative procedure in which the design variables are successively updated to obtain the optimal structural configuration. The strategy for updating the design variables based on the sensitivity information plays a crucial role in determining both the efficiency and the convergence of the optimization process. Over the years, various approaches have been proposed. In this study, we focus on three representative methods: the Method of Moving Asymptotes (MMA), the Reaction Diffusion Equation (RDE), and the Hamilton–Jacobi Equation (HJE), and provide an overview of their characteristics.

The Method of Moving Asymptotes

The Method of Moving Asymptotes (MMA) is arguably one of the most widely used optimization algorithms in contemporary topology optimization research. The reasons for its popularity lie in three main aspects: (i) its high level of generality, which allows it to be applied to a wide range of design problems, (ii) its relatively simple implementation, and (iii) its favorable convergence properties, leading to stable optimization histories. In the following, the basic concept of MMA is outlined, while details can be found in the original work (Svanberg, 1987).

Let us first consider the following general nonlinear constrained optimization problem:

$$\begin{aligned} \min \quad & f_0(\mathbf{x}) + a_0 z + \sum_{i=1}^m (c_i y_i + \frac{1}{2} d_i y_i^2) \\ \text{subject to} \quad & f_i(\mathbf{x}) - a_i z - y_i \leq 0, \quad i = 1, \dots, m \\ & \mathbf{x} \in X, \quad \mathbf{y} \geq \mathbf{0}, \quad z \leq 0, \end{aligned} \tag{1.13}$$

where

$$\begin{aligned} a_0 > 0, \quad a_i \geq 0, \quad c_i \geq 0, \quad d_i \geq 0 \\ c_i + d_i > 0, \quad a_i c_i > 0. \end{aligned} \tag{1.14}$$

As shown in the original paper (Svanberg, 1987), this formulation is highly general and applicable to a broad class of engineering problems, including topology optimization. In general, the objective function f_0 and the constraint functions $f_i (i = 1, \dots, m)$ are strongly nonlinear and often multimodal. For such non-convex problems, obtaining a global optimum at low computational cost is generally infeasible.

On the other hand, convex functions are highly desirable in optimization, because if convexity is guaranteed, global optimal solutions can be reached efficiently using numerical techniques such as gradient-based methods. The key strategy of MMA is therefore to approximate the original nonlinear functions by locally convex functions and to solve the resulting approximate optimization problem. This ensures that, at each optimization step, the subproblem remains computationally efficient, while the iterative updates of the solution gradually converge toward the optimum.

Concretely, at each iteration k , the original problem is replaced with the following convex programming problem:

$$\begin{aligned}
\min \quad & \tilde{f}_0^k(\mathbf{x}) + a_0 z + \sum_{i=1}^m (c_i y_i + \frac{1}{2} d_i y_i^2) \\
\text{subject to} \quad & \tilde{f}_i^k(\mathbf{x}) - a_i z - y_i \leq 0, & i = 1, \dots, m \\
& \alpha_j^k \leq x_j \leq \beta_j^k, & j = 1, \dots, n \\
& y_i \geq 0, & i = 1, \dots, m \\
& z \leq 0,
\end{aligned} \tag{1.15}$$

where each function $f_i (i = 0, \dots, m)$ is approximated by the following convex function:

$$\tilde{f}_i^k(\mathbf{x}) = \sum_{j=1}^n \left(\frac{p_{ij}^k}{u_j^k - x_j} + \frac{q_{ij}^k}{x_j - l_j^k} \right) + r_i^k, \quad i = 0, 1, \dots, m \tag{1.16}$$

This approximation has lower and upper asymptotes, l_j^k and u_j^k , within which the design variables are restricted to evolve. The parameters p , q , r are defined as follows:

$$\begin{aligned}
p_{ij}^k &= (u_j^k - x_j^k)^2 \left(A \max \left(\frac{\partial f_i}{\partial x_j}(\mathbf{x}^k), 0 \right) + B \max \left(-\frac{\partial f_i}{\partial x_j}(\mathbf{x}^k), 0 \right) + \frac{C}{x_j^{\max} - x_j^{\min}} \right) \\
q_{ij}^k &= (x_j^k - l_j^k)^2 \left(B \max \left(\frac{\partial f_i}{\partial x_j}(\mathbf{x}^k), 0 \right) + A \max \left(-\frac{\partial f_i}{\partial x_j}(\mathbf{x}^k), 0 \right) + \frac{C}{x_j^{\max} - x_j^{\min}} \right) \\
r_i^k &= f_i(\mathbf{x}^k) - \sum_{j=1}^n \left(\frac{p_{ij}^k}{u_j^k - x_j^k} + \frac{q_{ij}^k}{x_j^k - l_j^k} \right),
\end{aligned} \tag{1.17}$$

where A , B , and C are set to 1.001, 0.001, and 10^{-5} by default. Among these, r is determined such that the approximation passes through the current point, while p and q adjust the weighting between functions of type $1/(u_j^k - x_j)$ and $1/(x_j - l_j^k)$. Intuitively, when the sensitivity gradient is negative, increasing the design variable is expected to improve the objective function. In such cases, q is assigned a larger value while p takes a smaller one. Conversely, when the gradient is positive, the roles of p and q are reversed. This mechanism allows MMA to handle nonlinear problems robustly while relying only on local gradient information.

In addition, the step size of the design variable updates is controlled by the parameters α and β , defined as:

$$\begin{aligned}
\alpha_j^k &= \max \left\{ x_j^{\min}, l_j^k + 0.1(x_j^k - l_j^k), x_j^k - 0.5(x_j^{\max} - x_j^{\min}) \right\} \\
\beta_j^k &= \min \left\{ x_j^{\max}, u_j^k - 0.1(u_j^k - x_j^k), x_j^k + 0.5(x_j^{\max} - x_j^{\min}) \right\}.
\end{aligned} \tag{1.18}$$

These parameters prevent the design variables from changing excessively in a single step, thereby ensuring numerical stability of the algorithm.

The convex subproblem thus constructed is solved in an inner loop using Newton's method. The obtained solution is then used as the updated design variable, after which the state equation and adjoint equation are solved again to compute the sensitivities, and the process is repeated. As the optimization proceeds, the lower and upper asymptotes l and u gradually move closer to each other, leading the solution to converge.

In summary, the MMA method enables efficient and stable optimization even for highly nonlinear and multimodal topology optimization problems. Its usefulness has been widely recognized, and at present, it can be considered the most recommended method when constructing topology optimization algorithms. In this dissertation, MMA is adopted as the design variable update scheme in Chapters 2 and 4.

Reaction-diffusion equation

Next, we describe the method proposed by Yamada et al. (2010) based on the Reaction-Diffusion Equation (RDE).

One of the simplest sensitivity-based update schemes is the Optimality Criteria (OC) method. Owing to its simplicity, the OC method has been widely adopted in the early studies of topology optimization. However, this method suffers from significant drawbacks: its convergence strongly depends on the time step size dt , and when the material distribution alternates sharply between solid and void across discretized elements, discontinuities may arise in the sensitivity evaluation. This may lead to the breakdown of state-field analysis, ultimately causing the optimization process to fail.

As discussed in Section 1.2.1, topology optimization directly optimizes the material configuration, which inherently induces spurious discontinuities and local oscillations in the numerical solution. To address this issue, smoothing of design variables is of paramount importance. Among the smoothing techniques commonly employed in topology optimization, the averaging filter and the Helmholtz-type filter are the most prominent. The averaging filter is relatively simple to implement but is sensitive to boundary treatment and the choice of filter radius, whereas the Helmholtz-type filter provides a more robust PDE-based framework for smoothing the design field.

Against this background, Yamada et al. (2010) introduced a design update scheme formulated using the RDE as the following equation shows:

$$\begin{cases} \frac{\partial \rho}{\partial t} = - (C_{\text{diff}} J' - \tau \nabla^2 \rho) & \text{in } D \\ \frac{\partial \rho}{\partial n} = 0 & \text{on } \partial D, \end{cases} \quad \begin{matrix} (1.19a) \\ (1.19b) \end{matrix}$$

This approach simultaneously achieves two essential tasks within a single equation: the reaction term drives the design update in the sensitivity direction, while the diffusion term ensures smoothness and suppresses numerical instabilities. As a result, this method overcomes the shortcomings of the conventional OC approach and enables the stable evolution of physically meaningful topologies. In Chapter 3 of this study, we adopt this method to realize both sensitivity-driven updates and smoothing simultaneously.

Hamilton-Jacobi equation

This method was originally proposed by Wang et al., 2003; Allaire et al., 2004 and is often used in combination with the level set method described in Section 1.2.1. In this approach, the design variable is updated by solving the Hamilton–Jacobi equation, as shown below.

$$\frac{\partial \psi}{\partial t} + V|\nabla \psi| = 0, \quad (1.20)$$

Here, V denotes the shape sensitivity. The method modifies the structural configuration by moving the material–void boundary continuously, and thus it is categorized as a shape optimization rather than a topology optimization technique. Consequently, no new holes are generated during the optimization process. Nevertheless, by introducing a sufficient number of holes in the initial configuration, results similar to those obtained by topology optimization can be achieved.

One of the notable advantages of this method is that it naturally represents smooth boundary evolution and explicitly handles structural boundaries. On the other hand, during the optimization process, a reinitialization procedure is required, as shown below, and numerical stabilization necessitates the use of upwind discretization. Therefore, implementing this method from scratch demands both technical knowledge and a certain degree of trial and error.

$$\frac{\partial \psi}{\partial t} + \text{sign}(\psi_0)(|\nabla \psi| - 1) = 0 \quad \text{in } D \times R^+, \quad (1.21)$$

$$\psi(t = 0, x) = \psi_0(x) \quad \text{in } D, \quad (1.22)$$

The reinitialization step is introduced to maintain the gradient of the level set function at unity.

1.2.4 Summary

Naturally, the methods introduced above do not represent the full spectrum of topology optimization approaches. In recent years, new frameworks such as Topology Optimization of Binary Structures (TOBS) and Moving Morphable Components (MMC) have also been proposed. TOBS constrains the design variables to discrete 0–1 values, thereby avoiding the gray-scale issue inherent in density-based methods. On the other hand, MMC introduces explicit geometric features

(e.g., member length, width and position) as design variables, which facilitates better control over the design freedom and enables optimization with manufacturability considerations.

The choice of method should be made appropriately, depending on the characteristics and objectives of the optimization problem at hand. In this study, the density method was employed as the primary framework (with Chapter 4 deliberately exploiting gray-scale solutions), while the design variable updates were carried out using MMA in Chapters 2 and 4, and the reaction–diffusion equation in Chapter 3. For sensitivity analysis, the adjoint variable method based on continuum formulation was consistently used throughout.

1.3 Structural optimization method for the design of batteries

Structural optimization is a collective term for a group of methods that use a mathematical approach to determine the shape, dimensions, and material layout of a structure to maximize its performance under given constraints. It is broadly classified into sizing optimization, which treats dimensions (e.g., thickness, width) as design variables; shape optimization, which optimizes cross-sectional shapes; and topology optimization (TO), which finds the optimal distribution of material within a given design domain. After its development in the field of structural mechanics, its application has expanded to problems dominated by various physical phenomena, such as thermo-fluid and electrochemical systems. In the field of battery design, in particular, TO is gaining attention as a design tool that can resolve the complex trade-offs between multiple physical phenomena that determine performance—such as heat transport, mass transport, and electrochemical reactions—and create innovative structures that go beyond conventional concepts. This section provides an overview of previous research on battery design using TO, categorized into four areas: thermal management, flow batteries, solid oxide fuel cells, and lithium-ion batteries.

1.3.1 Thermal management

The performance and safety of lithium-ion batteries are highly dependent on temperature, making effective thermal management indispensable. To enhance the performance of thermal management components such as cooling plates and heat sinks for battery packs, structural optimization has long been applied. Early research focused on sizing optimization, where parameters like the width, height, and number of cooling channels were used as design variables (Rao and Wang, 2011). Methods such as response surface methodology and genetic algorithms were employed to design for the trade-off between cooling performance and pressure drop.

As an approach to overcome the limitations of such experience-based parameter design, in recent years, TO has been applied to enhance the performance of thermal management components

such as cooling plates and heat sinks for battery packs. Initial studies were primarily aimed at resolving the trade-off between cooling performance and pressure drop. Mo et al. (2021) applied TO to the design of a cooling plate for a battery thermal management system (BTMS) and created a topological structure that could reduce the maximum temperature by 2.3°C and the pressure drop by 47.9% compared to a traditional straight-channel plate. Similarly, Chen et al. (2022) numerically demonstrated that a TO-designed cold plate (TCP) significantly improves the temperature uniformity of the battery (up to a 41.88% reduction in temperature difference) compared to conventional rectangular-channel (RCP) and serpentine-channel (SCP) plates. These studies show that TO is effective for novel channel design that is not based on empirical rules.

Furthermore, to consider more realistic heat sources, research coupled with electrochemical models has been advanced. Guo et al. (2022a) accurately predicted the heat generation distribution of a battery using a 3D electrochemical-thermal coupled model and used it as the heat source for the TO of a cooling plate. This allowed them to derive optimal channel structures corresponding to different numbers and arrangements of inlets and outlets, the validity of which was also verified by experiments.

In recent years, applications have expanded beyond the optimization of simple heat transport characteristics to the design of multifunctional battery packs that simultaneously serve a structural role. Kambampati et al. (2021) applied level-set-based TO to the design of a load-carrying battery pack installed within an aircraft wing. In this study, a multiphysics problem coupling Darcy flow for the coolant, heat transfer, and thermo-elasticity was solved to perform a multi-objective optimization considering structural stiffness, the average temperature of the battery cells, and the coolant pressure drop. This demonstrated the potential for designing lightweight and highly efficient multifunctional structures that combine load-bearing and thermal management functions in a single component.

1.3.2 Flow battery

In improving the performance of redox flow batteries (RFBs), the design of flow fields and electrode structures that efficiently supply electrolyte to the porous electrode reaction sites is crucial. To address this challenge, sizing and shape optimization have been studied for many years, with design variables including flow channel dimensions (e.g., rib and channel widths in interdigitated flow fields) and electrode compression ratios (porosity) (Xu et al., 2013; Houser et al., 2017). These studies combine computational fluid dynamics (CFD) simulations with optimization algorithms to find parameters that maximize mass transport while minimizing pressure drop (Messaggi et al., 2018). As an approach to further increase design freedom beyond such parameter optimization, the application of TO in this field began with the design of flow fields external to the cell and has gradually deepened to the optimization of porosity distribution and microstructure within the electrode itself.

Yaji et al. (2018) conducted one of the pioneering studies applying TO to the flow field design of a vanadium redox flow battery (VRFB). The novelty of their work lies in modeling the electrode as a permeable porous medium, rather than the impermeable solid domain common in previous fluid TO studies, and incorporating a mass transfer coefficient dependent on the local velocity in a simplified 2D model. The resulting optimal shape resembled the interdigitated flow field, a configuration known for high performance, demonstrating that TO could autonomously derive effective designs. This approach was extended by Chen et al. (2019) to a more realistic 3D model incorporating Butler-Volmer electrochemical kinetics, which also confirmed the effectiveness of the interdigitated type. Lin et al. (2022) further expanded the design freedom from 2D channels to fully three-dimensional space, creating complex 3D flow fields that minimize electrical and pressure losses.

In parallel with these flow field designs, research on optimizing the electrode structure itself has advanced. Beck et al. (2021) proposed the concept of "architected electrodes" with spatially varying porosity. They used a homogenization method to calculate the effective properties of microstructure unit cells and applied them to a macroscopic continuum model to determine the optimal 3D porosity distribution for the entire electrode. This approach enables the electrode to function as its own integrated flow field, achieving significant performance improvements over conventional uniform porosity electrodes. Wolf et al. (2024) continued this focus, using an experimentally validated half-cell model to perform both TO for porosity distribution and shape optimization for the overall cell geometry. They found that lower porosity downstream of the inlet and higher porosity near the outlet is effective, and that a horizontal cell format is advantageous over the conventional vertical format under many conditions, challenging traditional cell design assumptions. Furthermore, Wang et al. (2025) proposed a new multi-objective optimization framework using the Independent Continuous Mapping (ICM) method and a genetic algorithm to optimize the 2D layout of microstructures, addressing issues of localized high pressure and reactant accumulation (dead zones).

1.3.3 Solid Oxide Fuel Cell (SOFC)

In the field of solid oxide fuel cells (SOFCs), structural optimization also contributes to performance enhancement. Design challenges in SOFCs include thermal stress due to high-temperature operation and complex electrochemical reaction fields. Sizing optimization approaches have targeted the mitigation of thermal stress and improvement of power generation efficiency, using design variables such as the thickness of electrode layers, parameters of composition profiles in functionally graded materials (FGMs) (Liu et al., 2012; Zhou et al., 2025), and dimensions of gas flow channels (Xu et al., 2016). Further advancing such research, TO, which aims for more unconstrained shape creation, also contributes to performance enhancement. Design challenges in SOFC research include thermal stress due to high-temperature operation and complex electrochemical reaction fields.

Iwai et al. (2011), in an early application of TO to SOFCs, focused on optimizing the shape of the cathode-electrolyte interface. Using the level-set method, they demonstrated that creating a wavy interface at the mesoscale (10–100 μm) increases the reaction area and improves current density by approximately 10% compared to a flat interface. This work was groundbreaking in that it targeted the geometry of the functional interface itself, rather than the macroscopic flow field or bulk porosity.

More recent research has expanded the scope to the design of flow channels considering the complex multiphysics phenomena of the entire cell. Goncalves et al. (2024) proposed an ingenious solution to the challenge of immense computational costs associated with 3D SOFC models. They developed a "design variable propagation" approach that couples a 2D optimization problem with a full 3D multiphysics model. In this method, the 2D design information is "propagated" into the 3D simulation space by solving a partial differential equation. This significantly reduces the complexity of the optimization calculation while retaining the advantages of a high-fidelity multiphysics model that accurately represents complex transport and electrochemical phenomena. As a result, they derived an optimal flow field that achieves a more uniform flow and current density distribution and a lower pressure drop compared to traditional serpentine and parallel channel designs.

1.3.4 Lithium ion battery

The performance of lithium-ion batteries is critically dependent not only on peripheral components like cooling systems but also on the microstructure of the porous electrodes where ions and electrons move and react. For a long time, sizing optimization based on electrochemical models has been vigorously pursued, using scalar parameters such as electrode thickness, active material particle size, and porosity as design variables (Sarawutanukul et al., 2021; Kench et al., 2024). These studies have employed methods like response surface methodology, genetic algorithms, and more recently, Bayesian optimization, to analyze the trade-off relationship between energy and power density (Ragone plots) and have achieved significant success in finding optimal parameters for specific objectives (Karaki et al., 2023; Chen and Zhang, 2024). To address this challenge, in recent years, the focus of TO has shifted from macroscopic channels and structures to the microscopic distribution of materials within the electrode itself.

In a pioneering study in this area, Roy et al. (2022) applied density-based TO directly to the design of porous electrode structures. Using a half-cell model, they targeted both steady-state Faradaic reactions (batteries) and transient charge/discharge cycles (supercapacitors) to minimize energy loss. They found that for material systems with low ionic conductivity, the optimization algorithm autonomously generates hierarchical structures composed of macropores and micropores, which exhibit superior performance to homogeneous electrodes.

Building on the success of this half-cell model, Li et al. (2024) extended the research to a full-cell model that handles the anode and cathode simultaneously. This was a significant step forward as it allows for the consideration of interactions between the two electrodes and the ion concentration distribution in the electrolyte. They introduced a boundary propagation method to distinguish the anode and cathode regions and coupled it with an electrochemical model that includes mass transport. Under ion-diffusion-limited conditions, the optimized electrodes exhibited complex, intertwined interdigital and 3D interlocked structures, achieving up to a 750% increase in energy storage capacity compared to conventional designs.

Efforts to explore the physical validity of optimization methods are also underway. Alizadeh et al. (2023) compared two different optimization strategies for electrode design: "current maximization at a given overpotential" and "overpotential minimization at a given current." Furthermore, they introduced entropy generation analysis as a metric to evaluate the irreversibility of the system and analyzed how thermodynamic losses change during the optimization process. Their results showed that the optimal structure has a tree-root-like branched configuration and that a less dissipative (higher performance) system corresponds to more uniform current density and entropy generation distributions.

On the other hand, a major challenge of TO is its enormous computational cost. To address this, Long et al. (2024) proposed a new design approach that bypasses TO itself, based on insights gained from analyzing TO results. They discovered a universal feature in various optimized electrode structures: the concentration distribution of reactants becomes nearly linear. By leveraging this finding, they developed a method to directly calculate the optimal porosity distribution of a porous electrode by solving the governing equations under the assumption of a linear concentration profile. This suggests the possibility of designing structures with performance comparable to TO at a low cost, without requiring large-scale iterative computations.

Furthermore, TO is also being applied to the design of multifunctional materials. Pejman and Najafi (2023) addressed the design of the electrolyte for a structural battery composite (SBC), which combines mechanical load-bearing capacity with energy storage functions. The structural battery electrolyte (SBE) requires both high ionic conductivity and high mechanical stiffness, which are conflicting properties. They constructed a multiphysics, multi-objective TO framework that couples the electrochemical, thermal, and structural fields, successfully visualizing this trade-off relationship as a Pareto front. This research demonstrates that TO is a powerful tool for optimizing the microstructure of materials under complex physical constraints.

1.3.5 Summary

This chapter provides a comprehensive review of the application of TO to battery design, highlighting its emergence as a powerful tool for creating innovative structures that surpass conventional, experience-based designs. The review is organized into four key areas, demonstrating the breadth and depth of TO's impact on enhancing battery performance and functionality. The application of TO is moving from single-physics, macroscale component design towards multi-physics, microscale structural optimization. By systematically navigating complex trade-offs between mass transport, heat transfer, electrochemical reactions, and mechanical properties, TO is proving to be an indispensable method for unlocking the next generation of high-performance battery designs.

Building on this review, the following chapters of this dissertation will present the TO method developed to address the problems discussed in Section 1.1.3. As outlined in Section 1.1.4, this method is based on continuous adjoint sensitivity analysis for the design of internal battery components.

Chapter 2

Topology optimization for the design of battery separators[†]

CONTENTS	
2.1 Introduction	25
2.2 Formulation	27
2.2.1 Design variable	27
2.2.2 Governing equations	30
2.2.3 Optimization problem	31
2.2.4 Sensitivity analysis	34
2.3 Implementation details	35
2.3.1 Optimization algorithm	35
2.4 Numerical examples	36
2.4.1 Verification of sensitivity derivation	37
2.4.2 Optimized configurations in 2D	38
2.4.3 Optimized configurations in 3D	44
2.4.4 Investigation for the objective function	47
2.5 Conclusions	49

2.1 Introduction

The separator in a lithium-ion battery is a thin-film component made of a porous material immersed in an electrolyte, and it plays a crucial role in battery performance and safety by separating the positive and negative electrodes. Separators require electrical insulation to prevent short circuits and high ion conductivity to facilitate ion movement (Yoshino, 2015). Furthermore, to ensure a path for the electrolyte, the separator must have sufficient stiffness to withstand the pressure from volume changes in the electrodes during charging and discharging, as well as external pressures. Additionally, a high tortuosity of the electrolyte path is required to suppress dendrite growth from metallic impurities. By satisfying these design requirements, separators enable the realization of high-performance and safe batteries.

[†]The work in this chapter has been published in *Journal of the Japan Society of Precision Engineering*. DOI: 10.2493/jjspe.91.791.

To improve the characteristics of separators while satisfying these design requirements, various efforts have been made in material and structural development (Choi and Kim, 2022). In material development, methods have been developed to combine polymer materials with heat-resistant ceramics to improve heat resistance and mechanical strength (Takaoka and Miyazu, 2014). Technologies have also been developed to enhance the affinity with the electrolyte and improve chemical stability by forming a coating on the separator surface (Kim et al., 2017).

In contrast, in structural development, a key challenge is the control of the size, distribution, and shape of the micropores within the porous separator. For example, it has been reported that arranging micropores uniformly can improve ion transport efficiency while reducing the risk of localized heat generation and short circuits (Huang et al., 2024; Liu and Chuan, 2021). Additionally, Shi et al. (Shi et al., 2015) developed a material structure with a three-dimensional porous framework possessing high heat resistance and tortuosity, and they experimentally confirmed that performance improves with structural changes.

As described above, there are numerous research reports on structural development aimed at improving the performance of battery separators, and the internal structure of the separator has a significant impact on battery performance. However, a fundamental improvement in performance is currently difficult because a design guideline based on physical laws has not yet been established.

Therefore, this study aims to establish a structural optimization design method for lithium-ion battery separators as a solution to this problem. Structural optimization is a method that derives an optimal structure based on mathematical and physical principles, enabling the design of an optimal separator structure that satisfies multiple design requirements. Among these methods, topology optimization is a promising new design approach for battery separators. Topology optimization is the most flexible method in structural optimization; by formulating the structural optimization problem as a material distribution problem, it allows for arbitrary shapes and morphologies, and has the potential to dramatically improve the performance of the target structure (Kondoh et al., 2024).

Since its proposal by Bendsøe and Kikuchi (1988), topology optimization has seen the development of methodologies for various physical phenomena, including thermo-fluid problems (Borrvall and Petersson, 2003; Matsumori et al., 2013) and electromagnetics (Wadbro and Berggren, 2006). In recent years, many studies have been reported on the construction of topology optimization design methods for batteries. Chen et al. (2019) and Lin et al. (2022) developed an optimal design method for redox flow batteries and constructed an optimal electrolyte flow path. Roy et al. (2022) used a parallelized analysis method to perform large-scale calculations and derive a more detailed optimal electrode structure in three dimensions. Li et al. (2024) constructed an optimization method capable of deriving the optimal electrode and electrolyte interface for a full cell. Furthermore, Ishida et al. (2024) developed a topology optimization method based on the homogenization method for all-solid-state batteries. As such, with the significant improvement in

the design freedom of structures due to recent advances in additive manufacturing, design methods for the internal geometry of batteries are attracting attention. Among these, to the best of our knowledge, a topology optimization design method for the microstructure of the separator, which is one of the important components of a battery, has not yet been proposed.

Therefore, this study aims to construct a topology optimization design method for designing a lithium-ion battery separator that considers both stiffness and ion conductivity while suppressing short circuits due to dendrite growth. While thinning the film of conventional porous separators has been difficult due to stiffness constraints, the proposed method is expected to enable further thinning of the separator by deriving an efficient structure while maintaining safety.

Hereinafter, Section 2.2 explains the formulation of this study. First, to efficiently represent the complex separator structure with a small number of design variables, a unit cell is defined, and a density function indicating the presence or absence of the separator material is introduced within it. Next, the governing equations for evaluating ion transport in the electrolyte in which the separator is immersed are described. Then, an optimization problem is formulated to derive the desired separator structure using an objective function based on concentration gradients, and the sensitivity is derived in a continuous system by applying the adjoint variable method. In Section 2.3, an optimization algorithm based on the gradient method is developed for the formulated optimization problem. Subsequently, in Section 2.4, numerical examples in two and three dimensions are presented to verify the proposed objective function and sensitivity derivation method. The effectiveness of this research is also demonstrated through a comparison of the solutions obtained from the optimization problem. Finally, Section 2.5 presents the conclusions of this study.

2.2 Formulation

In this chapter, the formulation is performed in both two and three dimensions. First, Section 2.2.1 defines the fixed design domain and the density function representing the presence or absence of the separator material. Then, Section 2.2.2 describes the formulation of the governing equations. Finally, Section 2.2.3 defines two optimization problems.

2.2.1 Design variable

Figure 2.1 shows schematics of two and three-dimensional lithium-ion battery and the fixed design domains in two and three dimensions. As shown in Fig. 2.1, a lithium-ion battery consists of electrodes, a separator, and an electrolyte. The separator, which is the subject of this study, is located between the positive and negative electrodes and is immersed in the electrolyte. In this study, a unit cell is introduced to efficiently represent the complex separator structure with a

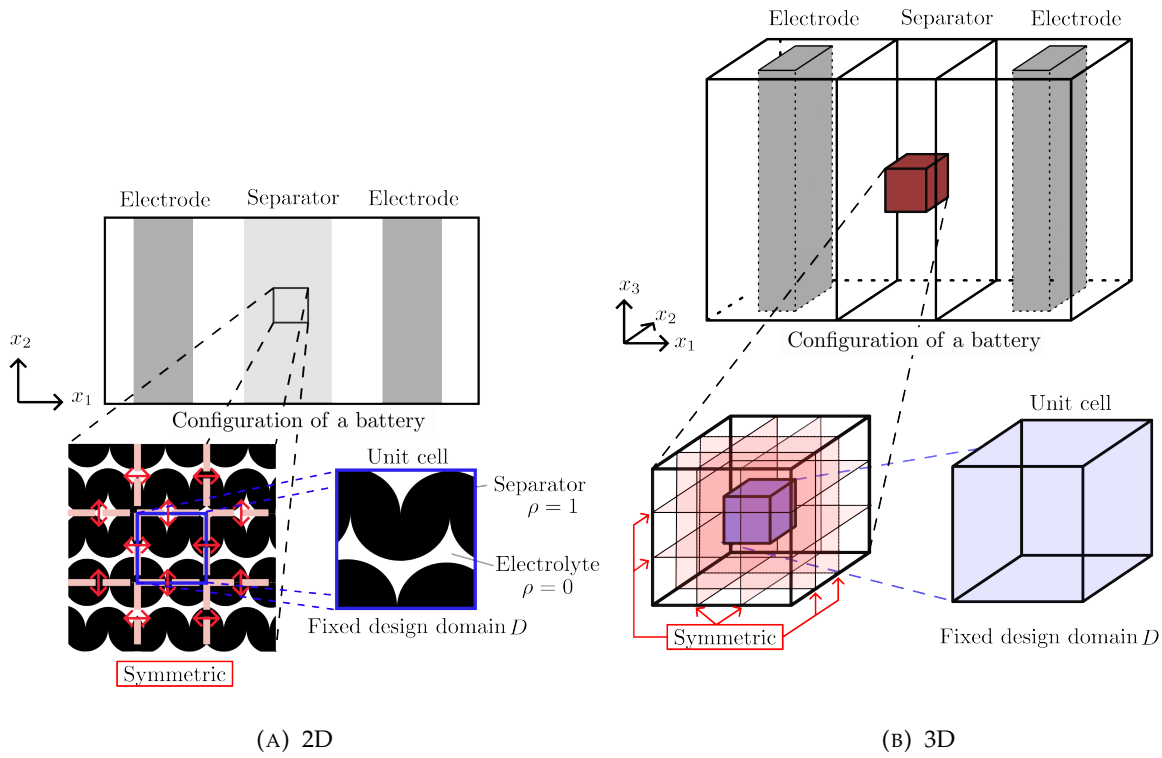


FIGURE 2.1: The configurations of a battery

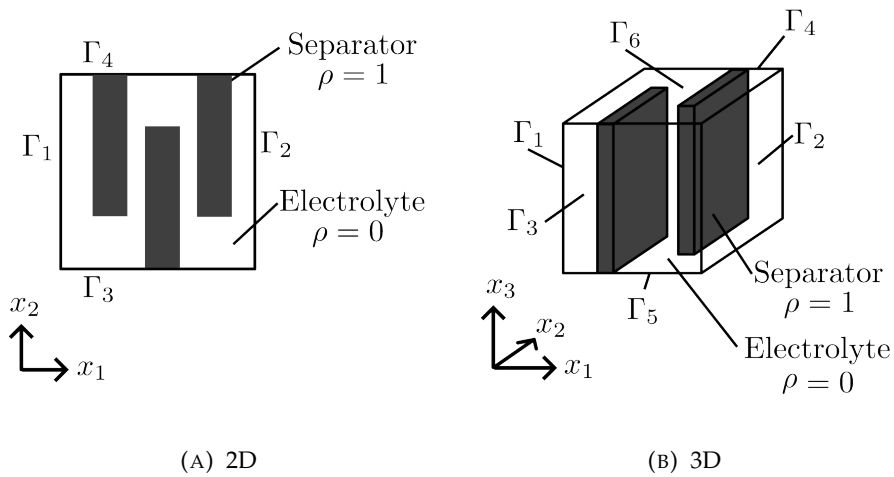


FIGURE 2.2: The definition of the unit cell

small number of design variables. That is, it is assumed that the unit cells within the separator exist symmetrically with respect to each face. Figure 2.2 show the unit cell domains in two and three dimensions, respectively. In this study, the unit cell is taken as the fixed design domain D , and its boundaries are defined as Γ_1 to Γ_4 in two-dimensional case and Γ_1 to Γ_6 in three dimensional case as shown in Fig. 2.2a and Fig. 2.2b, respectively. Next, the spatial coordinates within the unit cell are defined as follows.

$$\mathbf{x} = \begin{cases} (x_1, x_2) & \in D \subseteq R^2 \\ (x_1, x_2, x_3) & \in D \subseteq R^3, \end{cases} \quad (2.1)$$

Here, it is assumed that there are electrodes in the x_1 direction in both two and three dimensions. The design variable to represent the presence or absence of the separator within the fixed design domain D is defined by a density function $\rho(\mathbf{x})$ with a continuous value from 0 to 1. That is, the material distribution of the separator is represented using a function that takes the value 1 in the separator region and 0 in the region where the electrolyte exists.

$$\rho(\mathbf{x}) = \begin{cases} 1 & \text{separator} \\ 0 & \text{electrolyte} \end{cases} \quad (2.2)$$

Then, to avoid creating an overly complex structure within the separator, a smoothed density function $\bar{\rho}$ is obtained using a Helmholtz-type filter as shown in the following equation.

$$-R^2 \nabla \cdot \nabla \bar{\rho} + \bar{\rho} = \rho \quad \text{in } D, \quad (2.3)$$

$$\nabla \bar{\rho} \cdot \mathbf{n} = 0 \quad \text{on } \partial D, \quad (2.4)$$

Here, R is a constant proportional to the filter radius, and $\mathbf{n}$ is the outward unit normal vector. Also, ∂D is the boundary surrounding the fixed design domain D .

The smoothed density function $\bar{\rho}$ tends to have many intermediate values between 0 and 1, which increases the proportion of an intermediate state that is neither separator nor electrolyte and is difficult to interpret from an engineering perspective, making the structural boundaries unclear. To avoid this intermediate state and to achieve a clear boundary representation, the density function values are binarized using an approximate smoothed Heaviside function as shown in the following equation.

$$\rho_H = \frac{1}{2} \tanh \left(\beta \left(\bar{\rho} - \frac{1}{2} \right) \right) + \frac{1}{2}, \quad (2.5)$$

Here, β is a parameter that determines the steepness of the function.

2.2.2 Governing equations

This section describes the governing equations for solving the state field within the fixed design domain D . As mentioned earlier, this study focuses on the suppression of dendrite growth. Dendrites grow when impurity metals contained in the battery are oxidized at one electrode, become ions, pass through the electrolyte, and are reduced back to metal at the other electrode, with these metal deposits connecting and growing. That is, by deriving the transport path of ions passing through the separator region, the path along which dendrites grow can be determined. Therefore, this study focuses on the diffusion of ion concentration within the electrolyte.

The problem is generalized and formulated using dimensionless variables. First, a dimensionless ion concentration C is defined within the unit cell. As an equation to describe the ion concentration flux in the electrolyte, the Nernst-Planck equation, which consists of the gradient of ion concentration and the gradient of electric potential, can be mentioned (Doyle et al., 1993). For simplification, the electric potential is assumed to be constant within the microscale unit cell in this research, allowing the ion concentration flux $\mathbf{q}$ to be defined by the following equation:

$$\mathbf{q} = -K\nabla C, \quad (2.6)$$

Here, K is a distributed quantity representing the dimensionless diffusion coefficient, and it is defined as follows depending on the design variable ρ_H .

$$K(\rho_H) = (1 - \rho_H)K_{\max} + \rho_H K_{\min}. \quad (2.7)$$

$K_{\max}$ is the dimensionless ion conductivity of the electrolyte, and $K_{\min}$ is a sufficiently small value introduced to stabilize the calculation Allaire et al., 1997. In other words, in the region where the electrolyte exists ($\rho_H = 0$), $K = K_{\max}$, and in the region where the electrolyte does not exist, that is, where the separator exists ($\rho_H = 1$), K takes a small value, $K_{\min}$.

The governing equation for determining the dimensionless ion concentration C is shown below.

$$\nabla \cdot \mathbf{q} = 0 \quad \text{in } D, \quad (2.8)$$

$$C = 0 \quad \text{on } \Gamma_1, \quad (2.9)$$

$$C = 1 \quad \text{on } \Gamma_2, \quad (2.10)$$

$$\mathbf{q} \cdot \mathbf{n} = 0 \quad \text{on } \partial D \setminus \Gamma_{1,2}. \quad (2.11)$$

As mentioned in the previous section, it is assumed that the electrodes are placed so as to sandwich the separator in the x_1 direction. The difference in ion concentration, which is the driving force for ion movement, is caused by electrochemical reactions at the electrode-electrolyte interface. Therefore, to simulate the concentration difference originating from the region outside the unit cell, different values of Dirichlet boundary conditions are set on the boundaries Γ_1 and Γ_2 in the

direction of the electrodes, thereby simulating the ion concentration difference in the x_1 direction. Also, symmetry conditions are imposed on the other boundaries.

Furthermore, to evaluate the stiffness, which is one of the requirements for a battery separator, the displacement under compressive load in each direction is determined. That is, the dimensionless displacement vector $\mathbf{u}_i$ ($i = 1, 2, 3$) under compressive load in each direction is defined for the number of dimensions of the analysis domain. In this study, stiffness is considered only in the three-dimensional numerical examples. The strain tensor $\boldsymbol{\varepsilon}_i$ and stress tensor $\boldsymbol{\sigma}_i$ for each displacement are defined as follows.

$$\boldsymbol{\varepsilon}_i = \frac{1}{2}(\nabla \mathbf{u}_i + \nabla \mathbf{u}_i^T), \quad (2.12)$$

$$\boldsymbol{\sigma}_i = \frac{E}{1+\nu} \left(\boldsymbol{\varepsilon}_i + \frac{\nu}{1-2\nu} \text{tr} \boldsymbol{\varepsilon}_i \mathbf{I} \right), \quad (2.13)$$

Here, $\mathbf{I}$ is the identity matrix, and ν is the Poisson's ratio. Also, E is the dimensionless Young's modulus, and it depends on the design variable ρ_H as shown in the following equation.

$$E = (1 - \rho_H)E_{\min} + \rho_H E_{\max}, \quad (2.14)$$

Here, $E_{\max}$ is the dimensionless Young's modulus of the separator, and $E_{\min}$ is the dimensionless Young's modulus of the region where the separator does not exist, that is, where the electrolyte exists, and is given a sufficiently small value compared to $E_{\max}$. The respective displacements $\mathbf{u}_i$ when a compressive load is applied in each direction are derived from the following equations.

$$\nabla \cdot \boldsymbol{\sigma}_i = \mathbf{0} \quad \text{in } D, \quad (2.15)$$

$$\mathbf{u}_i = \mathbf{0} \quad \text{on } \Gamma_{2i-1}, \quad (2.16)$$

$$\boldsymbol{\sigma}_i \mathbf{n} = \mathbf{f}_i \quad \text{on } \Gamma_{2i}, \quad (2.17)$$

$$\boldsymbol{\sigma}_i \mathbf{n} = \mathbf{0} \quad \text{on } \partial D \setminus \Gamma_{2i-1, 2i}, \quad (2.18)$$

Here, when determining the displacement due to the compressive load in the i direction, the boundary Γ_{2i-1} is fixed. Also, a compressive load $\mathbf{f}_i$ is imposed on the boundary Γ_{2i} . $\mathbf{f}_i$ is a load vector with magnitude f in the x_1, x_2, x_3 directions, and is represented by the following equation.

$$\mathbf{f}_1 = \begin{bmatrix} -f \\ 0 \\ 0 \end{bmatrix}, \mathbf{f}_2 = \begin{bmatrix} 0 \\ -f \\ 0 \end{bmatrix}, \mathbf{f}_3 = \begin{bmatrix} 0 \\ 0 \\ -f \end{bmatrix}. \quad (2.19)$$

2.2.3 Optimization problem

The objective of this study is to derive a structure for a battery separator that prevents short circuits caused by dendrite growth. To prevent short circuits due to dendrite growth, it is effective to

promote ion diffusion in the plane perpendicular to the direction connecting the electrodes (x_1 direction) within the separator, thereby causing dendrites from impurity metals present in the battery to grow in the plane perpendicular to the x_1 direction and suppressing localized growth toward the electrodes. As shown in Fig. 2.3, in this study, the direction in which dendrites are not desired to grow within the separator region, that is, the plane perpendicular to the x_1 direction, is defined as plane A. That is, the magnitude of the ion concentration gradient in the direction of plane A is made to be large relative to the magnitude of the ion concentration gradient in all directions. As a result, as shown in Fig. 2.4, ions composed of impurities that cause dendrite growth will diffuse from the direction of plane A (the x_2 direction in the two-dimensional case) toward the dendrite growth point, causing the dendrite to grow in the direction of plane A and suppressing short circuits due to dendrite growth.

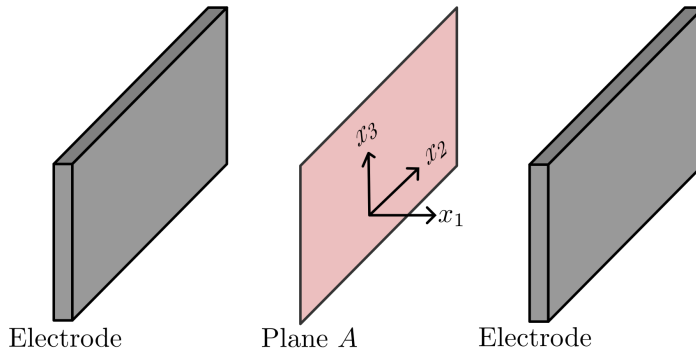


FIGURE 2.3: Definition of the plane A

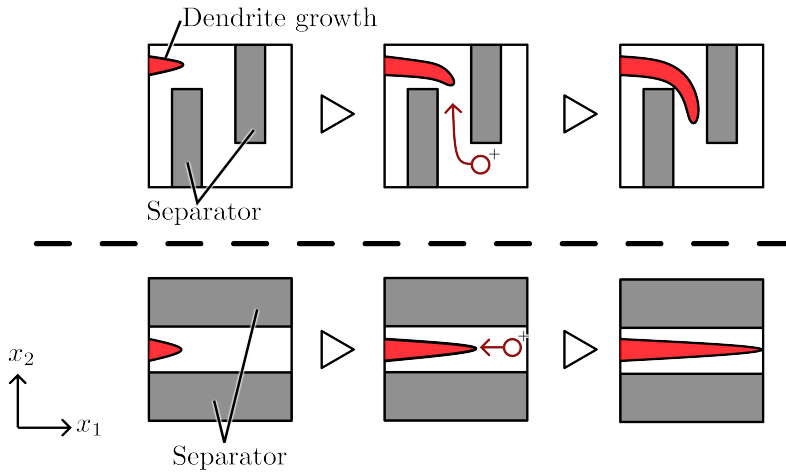


FIGURE 2.4: Schematics of dendrite growth in two-dimensions

In this study, an inequality-constrained optimization problem as shown in the following equations is defined.

[Optimization Problem]

$$\min_p \quad J = - \int_D \frac{K(A\nabla C) \cdot (A\nabla C)}{\nabla C \cdot \nabla C} dD, \quad (2.20)$$

$$s.t. \quad N_1 = 1 - \frac{\int K \nabla C \cdot \nabla C dD}{\tilde{N}_1} \leq 0, \quad (2.21)$$

$$N_2 = \frac{\int \varepsilon_1 : \sigma_1 dD}{\tilde{N}_2} - 1 \leq 0, \quad (2.22)$$

$$N_3 = \frac{\int \varepsilon_2 : \sigma_2 dD}{\tilde{N}_3} - 1 \leq 0, \quad (2.23)$$

$$N_4 = \frac{\int \varepsilon_3 : \sigma_3 dD}{\tilde{N}_4} - 1 \leq 0, \quad (2.24)$$

Here, the objective function is the function obtained by multiplying by -1 the ratio of the magnitude of the ion concentration gradient in the direction of plane A, which we want to maximize, to the magnitude of the ion concentration gradient. The magnitude of the ion concentration gradient in the direction of plane A can be evaluated using a matrix A composed of the unit basis vectors that form plane A, as shown in the following equation.

$$A = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \quad (\text{if } R^2), \quad \begin{bmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix} \quad (\text{if } R^3), \quad (2.25)$$

That is, by multiplying the concentration gradient vector by A , only the concentration gradient in the in-plane direction of plane A can be extracted. The effectiveness of the objective function will be verified in the numerical examples.

As will also be mentioned in the numerical examples, if the defined optimization problem is executed in three dimensions without considering the constraints, the obtained optimal structure is a solution that is not connected anywhere. As a method to suppress floating solids, the Virtual Temperature Method (VTM) has been proposed (Liu et al., 2015; Yamada and Noguchi, 2022; Sasaki et al., 2024). In addition, Kozuka et al. (Kozuka et al., 2023) identified floating solids based on image processing and suppressed them in the optimal structure by giving them a virtual energy. Although these methods can suppress floating solids, adding constraints when constructing a design method for a separator with many design requirements may narrow the feasible region. On the other hand, in problems where a material distribution that includes regions without material is obtained as the optimal solution, such as eigenvalue maximization problems (Allaire and Jouve, 2005) and linear buckling load maximization problems (Ferrari and Sigmund, 2019), a solution is sought by introducing stiffness constraints or concentrated masses. Therefore, in this study, by considering stiffness, which is one of the design requirements of the separator, as a constraint, we aim to simultaneously satisfy the design requirements and suppress floating solids. In addition, ion conductivity, another design requirement for battery separators, is also added to the constraints. Here, $\tilde{N}_1$ included in equation (2.21) is the lower limit value of the ion flux constraint,

and $\tilde{N}_2$, $\tilde{N}_3$, and $\tilde{N}_4$ included in equations (2.22)-(2.24) are the upper limit values of the respective stiffness constraints.

2.2.4 Sensitivity analysis

Next, the design sensitivity, which is the sensitivity with respect to the objective function, is derived using the continuous adjoint variable method. The derivation of the sensitivity regarding the constraints is left to previous research (Kawamoto et al., 2011). First, a Lagrangian is defined as shown in the following equation.

$$\mathcal{L} = J + \int_D \tilde{C} \nabla \cdot (-K \nabla C) \, dD, \quad (2.26)$$

Here, $\tilde{C}$ is called the adjoint variable and is a variable distributed within the fixed design domain. It should be noted that the governing equations for the displacement, which is a dependent variable not included in the objective function, are not included in the Lagrangian because they become zero in the functional differentiation procedure with respect to the concentration field C described later. This Lagrangian is functionally differentiated with respect to the design variable ρ to derive the design sensitivity. As shown by Kawamoto et al. (Kawamoto et al., 2011), to obtain the functional derivative of the Lagrangian with respect to the design variable ρ , one can perform functional differentiation with respect to the smoothed variable $\bar{\rho}$ and then apply the same filter to the obtained sensitivity.

The change in the Lagrangian $\delta \mathcal{L}$ when the density function $\bar{\rho}$ changes by a small amount $\delta \bar{\rho}$ is expressed as the sum of the partial variation due to the state variable C , $\delta_C \mathcal{L}$, and the partial variation due to the density function $\bar{\rho}$, $\delta_{\bar{\rho}} \mathcal{L}$.

$$\delta \mathcal{L} = \delta_{\bar{\rho}} \mathcal{L} + \delta_C \mathcal{L}. \quad (2.27)$$

By finding an adjoint variable $\tilde{C}$ such that $\delta_C \mathcal{L} = 0$, only the partial variation due to the density function $\bar{\rho}$, $\delta_{\bar{\rho}} \mathcal{L}$, contributes to the variation $\delta \mathcal{L}$. Using partial integration and Green's theorem, $\delta_C \mathcal{L}$ can be transformed into the following equation.

$$\begin{aligned} \delta_C \mathcal{L} = & \int_D \nabla \cdot 2K \left(\frac{(\nabla C \cdot \nabla C) A \nabla C}{(\nabla C \cdot \nabla C)^2} - \frac{(\nabla C \cdot A \nabla C) \nabla C}{(\nabla C \cdot \nabla C)^2} \right) \delta C \, dD \\ & + \int_D \nabla \cdot \tilde{\mathbf{q}} \, \delta C \, dD - \int_{\partial D \setminus \Gamma_{1,2}} \tilde{\mathbf{q}} \cdot \mathbf{n} \, \delta C \, d\Gamma \\ & + \int_{\Gamma_{1,2}} \tilde{C} \, \delta \mathbf{q} \cdot \mathbf{n} \, d\Gamma, \end{aligned} \quad (2.28)$$

Here, the flux of the adjoint field $\tilde{\mathbf{q}}$ is defined as follows:

$$\tilde{\mathbf{q}} = -K \nabla \tilde{C}. \quad (2.29)$$

Therefore, from the condition $\delta_C \mathcal{L} = 0$, the adjoint equation governing the adjoint variable $\tilde{C}$ is derived as follows:

$$\nabla \cdot \tilde{q}_3 = -\nabla \cdot 2K \left(\frac{(\nabla C \cdot \nabla C) A \nabla C}{(\nabla C \cdot \nabla C)^2} - \frac{(\nabla C \cdot A \nabla C) \nabla C}{(\nabla C \cdot \nabla C)^2} \right) \quad \text{in } D, \quad (2.30)$$

$$\tilde{C} = 0 \quad \text{on } \Gamma_{1,2}, \quad (2.31)$$

$$\tilde{q} \cdot \mathbf{n} = 0 \quad \text{on } \partial D \setminus \Gamma_{1,2}. \quad (2.32)$$

Therefore, the second term on the right-hand side of equation (2.27) becomes zero, and the right-hand side can be transformed as follows:

$$\delta \mathcal{L} = \delta_{\bar{\rho}} \mathcal{L}, \quad (2.33)$$

$$= \int_D \frac{\partial K}{\partial \rho_H} \left(\nabla C \cdot \nabla \tilde{C} - \frac{(A \nabla C) \cdot (A \nabla C)}{\nabla C \cdot \nabla C} \right) \frac{\partial \rho_H}{\partial \bar{\rho}} \delta \bar{\rho} \, dD, \quad (2.34)$$

$$:= \int_D S \, dD, \quad (2.35)$$

Here, S is a variable distributed in the fixed design domain and is the sensitivity of the objective function with respect to the smoothed design variable $\bar{\rho}$.

Finally, as shown in the following equation, the design sensitivity S_f is obtained by applying the same smoothing as in equations (2.3)–(2.4) to S (Kawamoto et al., 2011).

$$-R^2 \nabla \cdot \nabla S_f + S_f = S \quad \text{in } D, \quad (2.36)$$

$$\nabla S_f \cdot \mathbf{n} = 0 \quad \text{on } \partial D. \quad (2.37)$$

The verification of the sensitivity derivation is performed in Section 2.4.1, the effectiveness of the sensitivity derivation is shown in Section 2.4.2, and the details of derivation is summarized in Appendix A.1.

2.3 Implementation details

2.3.1 Optimization algorithm

This chapter explains the algorithm of the proposed topology optimization method. Figure 2.5 shows the flowchart. First, an initial value is given to the design variable ρ . Then, the governing equations are solved based on the distribution of that design variable. Next, using the obtained state variables, the values of the objective function and constraint functions are evaluated. Then,

the convergence of the optimization problem is determined based on the following equation.

$$\left\| \frac{J_r - J_{r-1}}{J_{r-1}} \right\| \leq \omega, \quad (2.38)$$

Here, r is the number of optimization iterations, and ω is a threshold for the convergence criterion. If this condition is satisfied, the optimization calculation is judged to have converged, and the optimization is terminated. On the other hand, if the condition is not satisfied, the adjoint equations are solved to calculate the sensitivity. Then, based on the derived sensitivity, the design variables are updated using the Method of Moving Asymptotes (MMA) (Svanberg, 1987). After that, the governing equations are solved using the updated design variables, and the above procedure is repeated until the objective function converges.

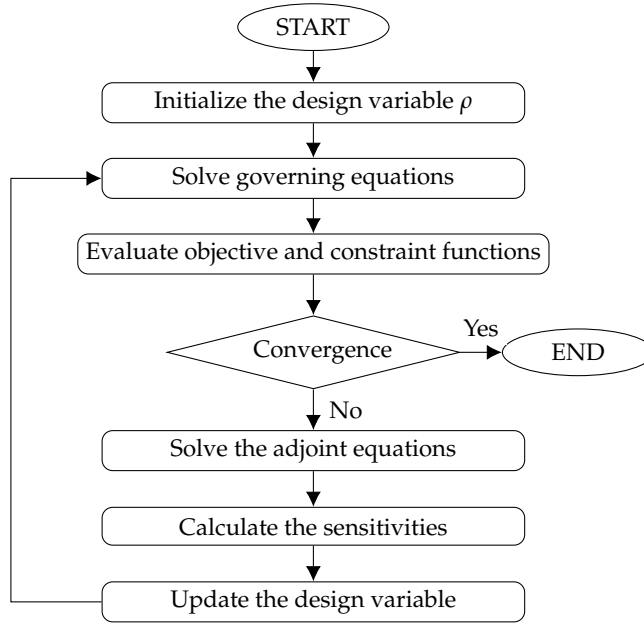


FIGURE 2.5: Optimization flow chart.

2.4 Numerical examples

Figure 2.6 shows the fixed design domain and mesh used in this numerical example. As shown in Fig. 2.6a, in the two-dimensional numerical example, a square with a side length of 0.5 was used as the fixed design domain, and the space was discretized using quadrilateral elements with a side length h of 0.001. The left, right, bottom, and top surfaces were designated as Γ_1 , Γ_2 , Γ_3 , and Γ_4 , respectively. On the other hand, in the three-dimensional numerical example, as shown in Fig. 2.6b, a cube with a side length of 0.5 was used as the fixed design domain, and it was discretized using hexahedral elements with a side length h of approximately 0.0125. The bottom, top, left, right, front, and back surfaces were designated as Γ_1 , Γ_2 , Γ_3 , Γ_4 , Γ_5 , and Γ_6 , respectively. The value of R

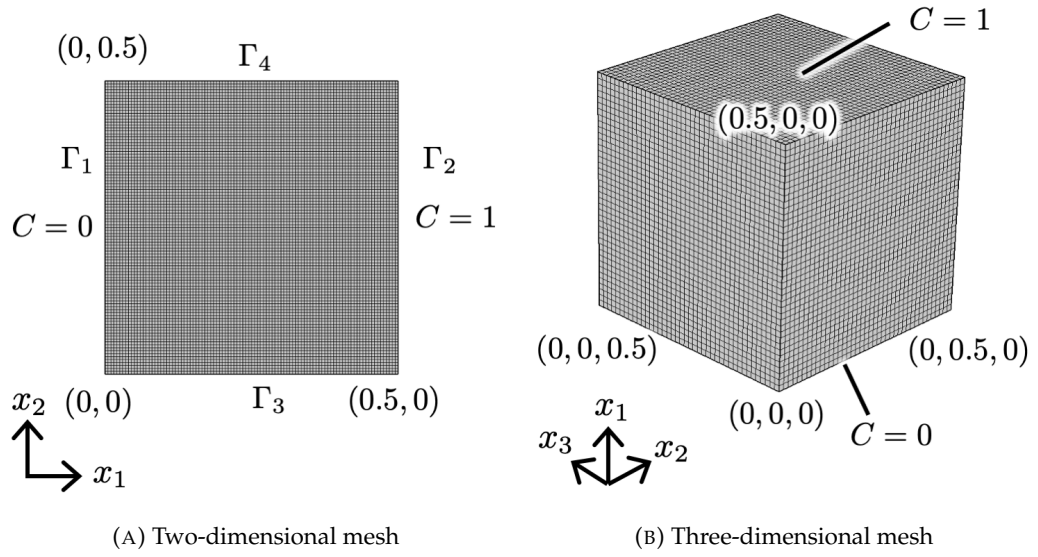


FIGURE 2.6: The definition of the unit cells and discretization

used in Eqs. (2.3) and (2.36) was 0.01 (equivalent to 10 elements) in the two-dimensional numerical example, and 0.015 (equivalent to 1.2 elements) in the three-dimensional numerical example. Also, $K_{\max}$ and $E_{\max}$ were set to 1, $K_{\min}$ and $E_{\min}$ were set to 0.001, and the Poisson's ratio ν was set to 0.3. Furthermore, the magnitude of the compressive load f was set to 0.25, and the threshold ω used for the optimization convergence criterion was set to 0.001.

2.4.1 Verification of sensitivity derivation

In this section, the validity of the design sensitivity S_f , derived by continuous adjoint sensitivity analysis in Section 2.2.4, is verified. Although there are several methods for verifying the sensitivity derivation, this study uses a comparison with the sensitivity $S_{f,\text{auto}}$ obtained by discrete adjoint sensitivity analysis. The design sensitivity S_f derived by continuous adjoint sensitivity analysis is based on functional differentiation and indicates the rate of change of the objective function when the design variable at a certain point changes. On the other hand, the sensitivity $S_{f,\text{auto}}$, which is derived using the global element stiffness matrix, indicates the rate of change of the objective function when the design variable at a certain node in the discretized analysis domain changes. Therefore, by multiplying the design sensitivity S_f obtained in the continuous system by the area over which the change in the design variable at each node has an effect, it should match the sensitivity $S_{f,\text{auto}}$ derived in the discrete form.

Figure 2.7 shows the distribution of the given design variables, the distribution of the design sensitivity S_f for each distribution of the design variables multiplied by the area of the discrete element h^2 (or $h^2/2$ at the boundary), and the distribution of the discrete adjoint sensitivity $S_{f,\text{auto}}$.

Figs. 2.7a, 2.7e and 2.7i show the distribution of the given design variables. These material distributions are the initial values used in the optimization problem shown in Section 2.4.2. Here, β was set to 6. In this study, the discrete adjoint sensitivity was derived using the sensitivity module of COMSOL Multiphysics. Figs. 2.7b, 2.7f and 2.7j show the discrete sensitivities, while Figs. 2.7c, 2.7g and 2.7k show the distributions of the derived continuous sensitivities multiplied by the area of influence of each node. It can be seen that the trends of the discrete and continuous sensitivity distributions are consistent for each condition. For a numerical comparison, Figs. 2.7d, 2.7h and 2.7l show the distribution of the $L2$ norm of the difference between the discrete sensitivity and the continuous sensitivity multiplied by the area of influence of each node. Observing the scale of the $L2$ norm distribution, it can be confirmed that in all conditions, it is less than one-tenth of the scale of the sensitivity. These results indicate that the two sensitivities are in good agreement, demonstrating the validity of the sensitivity derivation.

Here, the reason why the two distributions do not perfectly match can be attributed to implementation issues. The difference between discrete and continuous sensitivities lies in whether the adjoint variable method is applied before or after discretization to derive the sensitivity. Therefore, as shown in Figs. 2.7d, 2.7h and 2.7l, it is difficult to completely match the two distributions in regions where material is present or where the material distribution changes, due to the different timing of numerical integration and the difficulty of perfectly matching the numerical integration methods.

2.4.2 Optimized configurations in 2D

In this section, three two-dimensional numerical examples from #1 to #3 are shown. In the following numerical examples, β was linearly changed from 2 to 6 over 100 steps during the optimization process.

Figure 2.8 shows the results obtained by executing the proposed optimization problem without considering constraints for different initial values. As initial values, four types of material distributions as shown in Figs. 2.8a, 2.8d and 2.8g were given. The optimal structures obtained for each initial value are shown in Figs. 2.8b, 2.8e and 2.8h, and the dimensionless concentration distributions and ion flux vectors $\mathbf{q}$ in those optimal structures are shown in Figs. 2.8c, 2.8f and 2.8i. The horizontal direction in the figures is the x_1 direction.

From the optimal structure and the ion flux vector obtained in #1, it can be seen that the separator structure is distributed so as to curve the electrolyte path. Specifically, it is a structure that promotes dendrite growth in the direction perpendicular to the direction where the electrodes exist (x_2 direction) and suppresses growth in the x_1 direction. Also, comparing #2 and #3, it can be seen that the number of long separators in the x_2 direction included in the optimal structure strongly depends on the initial structure. This is thought to be because the proposed method is

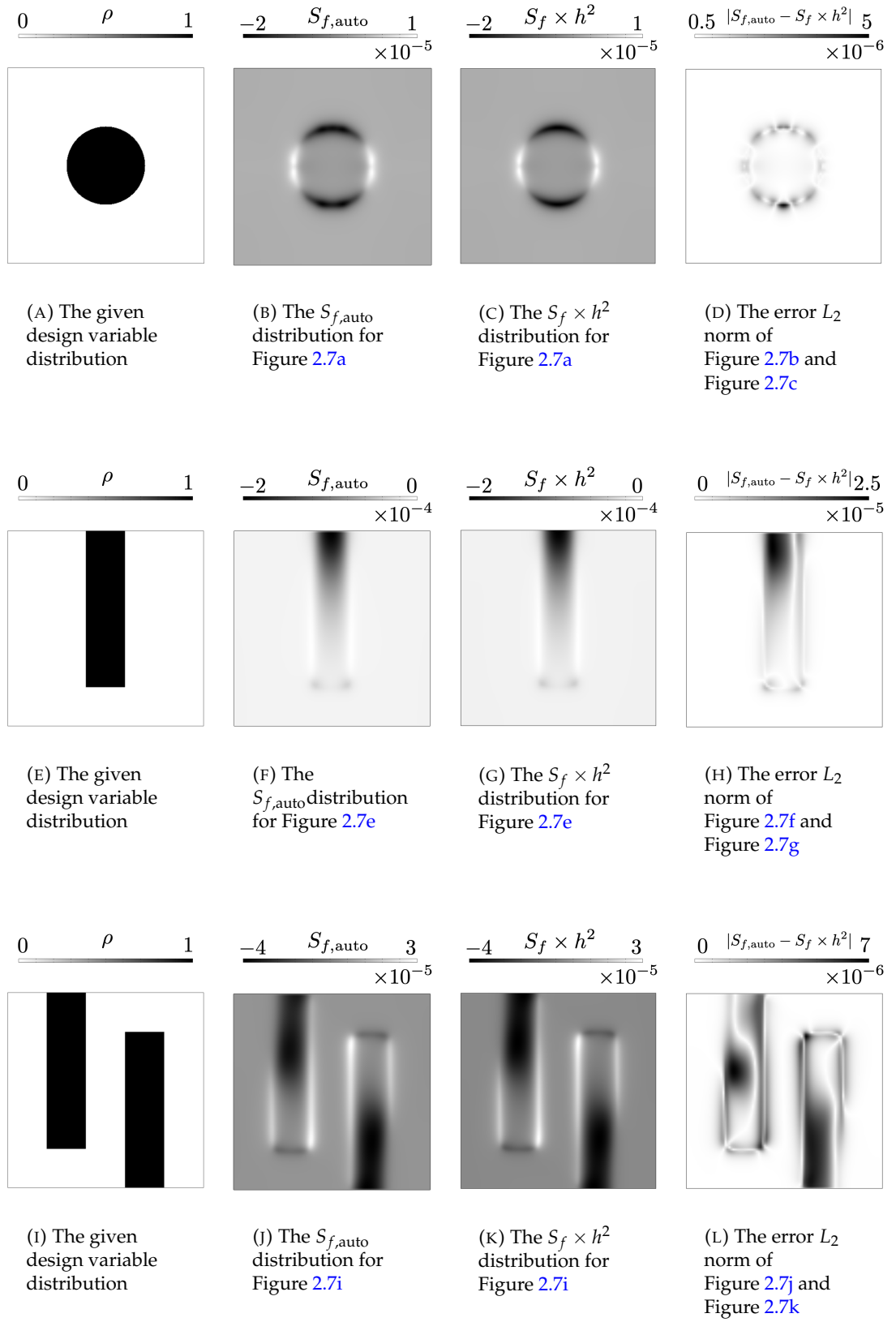


FIGURE 2.7: The comparison of the discrete sensitivity distribution $S_{f,\text{auto}}$ and the distribution of the product of the continuous sensitivity and the element area $S_f \times h^2$ for three types of design variable distributions

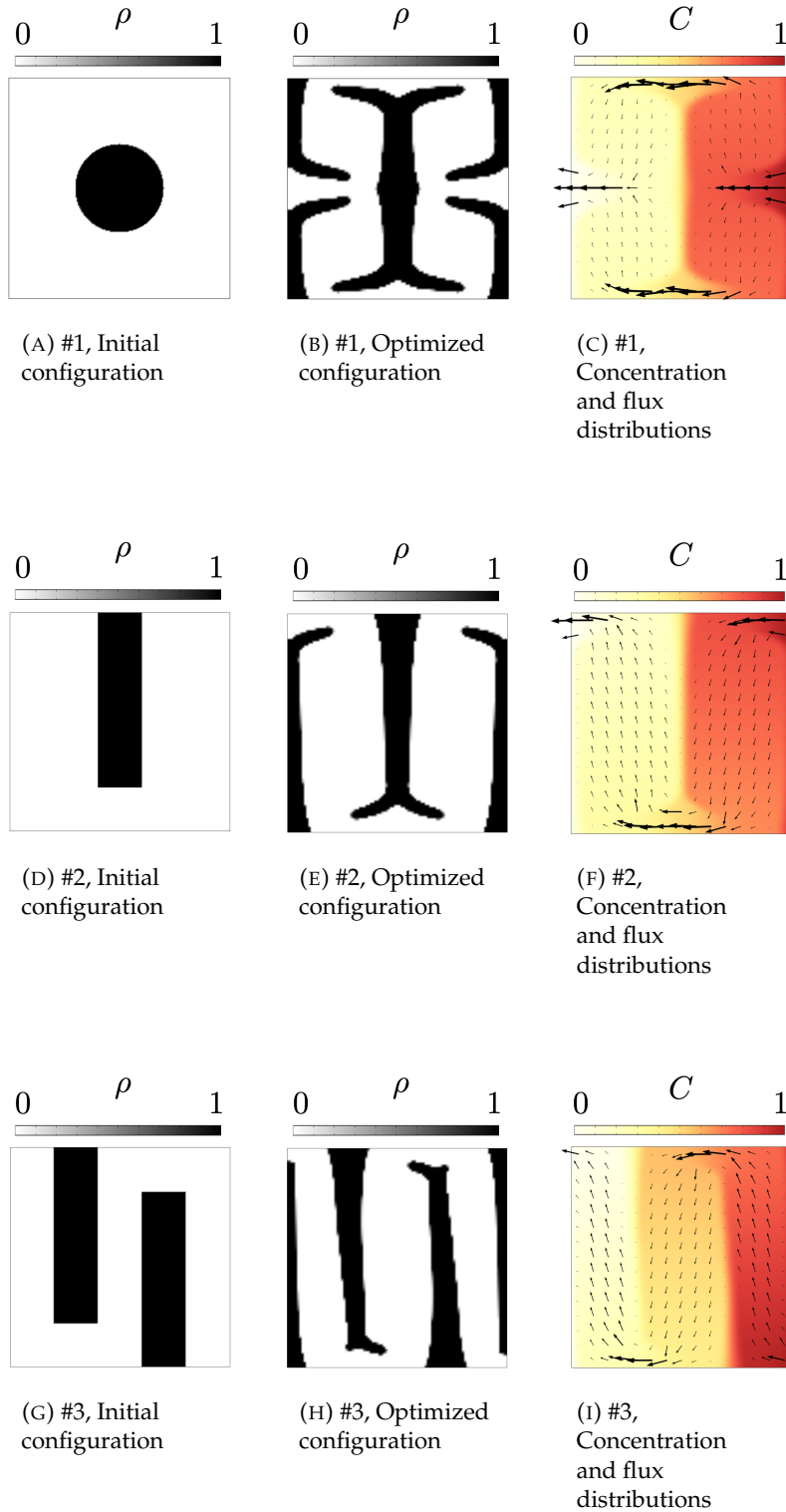


FIGURE 2.8: The configurations of the initial and optimized design variables, and the distributions of the concentration and flux (black arrow) for #1, #2, and #3

based on a gradient method, so the solution is trapped in a local optimum. However, in either case, it can be confirmed that the tip of the separator extends in the x_1 direction, which tends to curve the electrolyte path.

Figure 2.9 plots the history of the objective function values during the optimization process for conditions #1 to #3. As mentioned earlier, in this study, β was linearly changed from 2 to 6 over 100 steps during the optimization process. Therefore, the objective function value continues to change until the number of optimization iterations reaches 100 for all conditions. After 100 iterations, there is almost no change in the objective function, and the optimization terminates when the condition shown in Eq. (2.38) is satisfied. In all conditions, it was confirmed that the objective function decreased and converged using the proposed method.

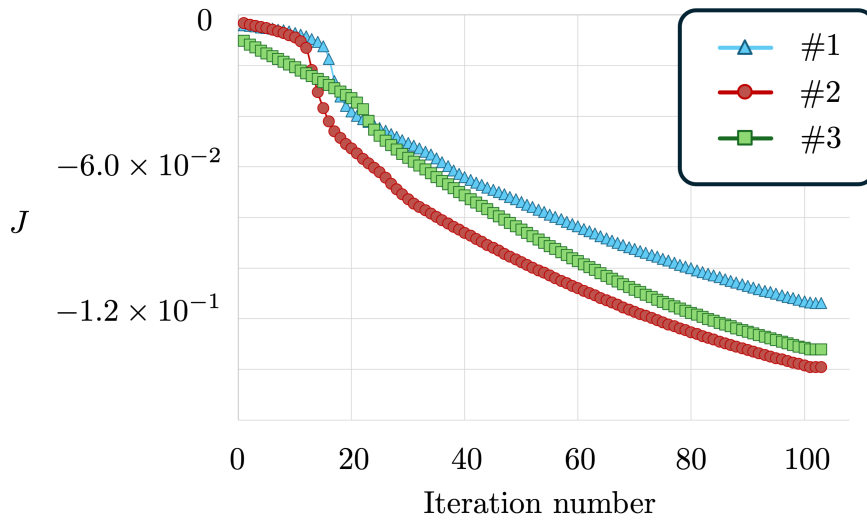


FIGURE 2.9: Objective function histories for #1 to #3

Next, the effectiveness of the proposed objective function is verified. One of the indicators for evaluating the degree of suppression of dendrite growth is tortuosity. Therefore, the tortuosity of the optimal structure is determined. As described in Section 2.2.1, the proposed method assumes that the unit cells are arranged symmetrically based on each boundary. However, as shown in Eqs. (2.8) and (2.11), although symmetry conditions are given for the boundaries in the direction of plane A, symmetry conditions are not applied to the boundaries in the x_1 direction. Therefore, the obtained optimal structure is arranged as assumed, and the tortuosity is measured by comparing the flux with a porous body having a specific tortuosity.

Figure 2.10a shows the calculation domain for recalculation. The recalculation domain Ω has a height of 1.5 and a width of 3.5, and consists of a region where the electrolyte exists, Ω_e , and a region where the separator exists, Ω_s . The left, right, top, and bottom boundaries of the calculation domain Ω are denoted as $\Gamma_{e,1}$, $\Gamma_{e,2}$, $\Gamma_{e,3}$, and $\Gamma_{e,4}$, respectively. Then, a dimensionless concentration

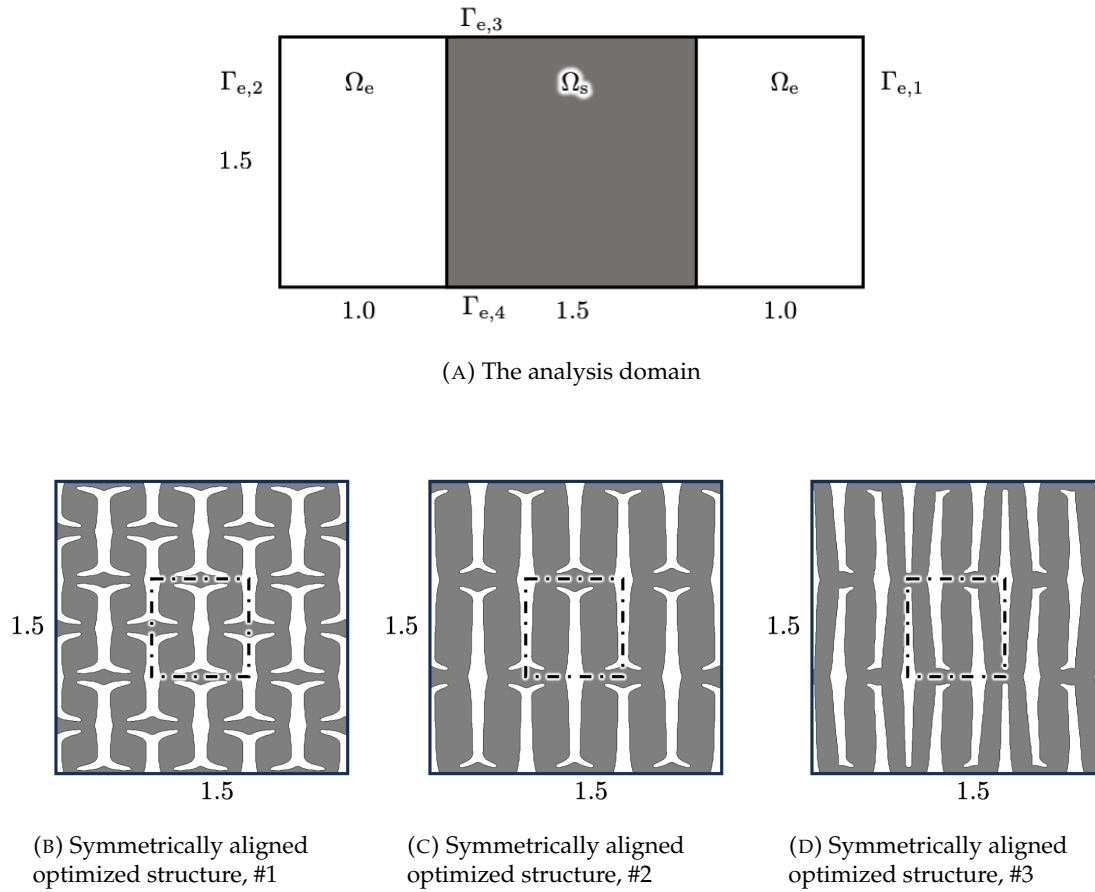


FIGURE 2.10: The analysis domain for evaluating the tortuosity factor of optimized separator configurations (a), and symmetrically aligned optimal structures with only the electrolyte region extracted (b–d)

$\bar{C}$ is defined in the domain Ω , and the flux $\bar{q}$ is formulated as shown in the following equation.

$$\bar{q} = -\bar{K}\nabla\bar{C}, \quad (2.39)$$

Here, $\bar{K}$ is the dimensionless ion conductivity. First, we show $\bar{K}$ when the separator is assumed to be made of a porous material. As shown in the following equation, $\bar{K}$ is defined to be 1 in the region where only the electrolyte exists, Ω_e , and ε/τ in the region where the porous separator exists, Ω_s .

$$\bar{K} = \begin{cases} 1 & \text{in } \Omega_e \\ \frac{\varepsilon}{\tau} & \text{in } \Omega_s, \end{cases} \quad (2.40)$$

Here, ε and τ are the porosity and tortuosity of the porous separator, respectively.

Next, the method for creating the calculation domain to verify the obtained optimal structure is explained. As shown in Figs. 2.10b–d, the optimal structures obtained in #1 to #3 were arranged symmetrically with respect to the boundaries of each unit cell. The unit cell has a side length of 0.5, and by arranging these in a 3×3 array, the size is adjusted to be the same as that of the porous separator. In other words, by swapping the region where the porous separator existed with the region of the symmetrically arranged optimal structure, analysis under the same conditions becomes possible. Also, the contour line where the density function ρ is 0.5 is defined as the boundary, and only the region where the electrolyte exists is defined as Ω_s . Therefore, the region where the separator itself exists is excluded from the calculation domain. As a result, the dimensionless ion conductivity $\bar{K}$ in the recalculation for the optimal structure is set to 1 in the entire domain, as shown in the following equation.

$$\bar{K} = 1 \quad \text{in } \Omega_e \cup \Omega_s. \quad (2.41)$$

Then, based on the ion flux defined above, the following diffusion equation is solved.

$$\nabla \cdot \bar{q} = 0 \quad \text{in } \Omega, \quad (2.42)$$

$$\bar{C} = 0 \quad \text{on } \Gamma_{e,1}, \quad (2.43)$$

$$\bar{C} = 1 \quad \text{on } \Gamma_{e,2}, \quad (2.44)$$

$$\bar{q} \cdot \mathbf{n} = 0 \quad \text{on } \Gamma_{e,3,e,4}, \quad (2.45)$$

Here, Dirichlet boundary conditions of 0 and 1 are set on $\Gamma_{e,1}$ and $\Gamma_{e,2}$, respectively, and a zero Neumann boundary condition is set on $\Gamma_{e,3}$ and $\Gamma_{e,4}$. For the obtained $\bar{C}$, the magnitude of the ion flux flowing out from the left face is calculated using the following equation.

$$Q = \int_{\Gamma_{e,1}} \bar{q} \cdot \mathbf{n} \, d\Gamma. \quad (2.46)$$

TABLE 2.1: The volume fraction of electrolyte regions within the unit cells, and tortuosity factor of the optimized separator structures obtained from the condition, #1, #2, and #3

No.	ε	τ
#1	0.660	9.87
#2	0.736	13.3
#3	0.702	15.3

The procedure for estimating the tortuosity of the optimal structure is described. First, for the continuous separator structure generated from the obtained optimal structure, Eqs. (2.42) and (2.45) are solved, and the magnitude of the ion flux flowing out from the left face, Q , is calculated using Eq. (2.46). Next, the porosity of the optimal structure (the area occupied by the electrolyte divided by the total area of the unit cell) is calculated. Then, the calculated porosity of the optimal structure is substituted for the porosity of the porous separator, ε , in Eq. (2.40), and while varying the tortuosity of the porous separator, τ , the tortuosity τ that matches the ion flux flowing out from the left face, Q , is derived.

Table 2.1 shows the porosity of the optimal structures obtained in #1 to #3, and the tortuosity obtained by recalculation. As can be seen from the table, the tortuosity of the obtained optimal structures was 9.8 or higher. On the other hand, since the tortuosity of a normal porous separator is set to about 1.0 to 2.0 (Waki et al., 2012), it was confirmed that the tortuosity is significantly improved by the proposed method.

2.4.3 Optimized configurations in 3D

Table 2.2 shows the conditions of the three-dimensional numerical examples presented in this section and the objective function values of the obtained optimal solutions. First, in #4, the optimization problem is solved without considering constraints. Next, in #5, the optimization problem is solved considering only the stiffness constraint. Finally, in #6, in addition to the stiffness constraint, an inequality constraint related to ion conductivity is considered. The initial structure for these numerical examples was a sphere with center coordinates (0.25, 0.25, 0.25) and radius 0.20.

TABLE 2.2: The set constraint values and the final objective function values for each condition, from #4 to #6

No.	$\tilde{N}_1$	$\tilde{N}_2$	$\tilde{N}_{3,4}$	J
#4	—	—	—	-0.0670
#5	—	0.1	1	-0.0202
#6	0.15	0.1	1	-0.0187

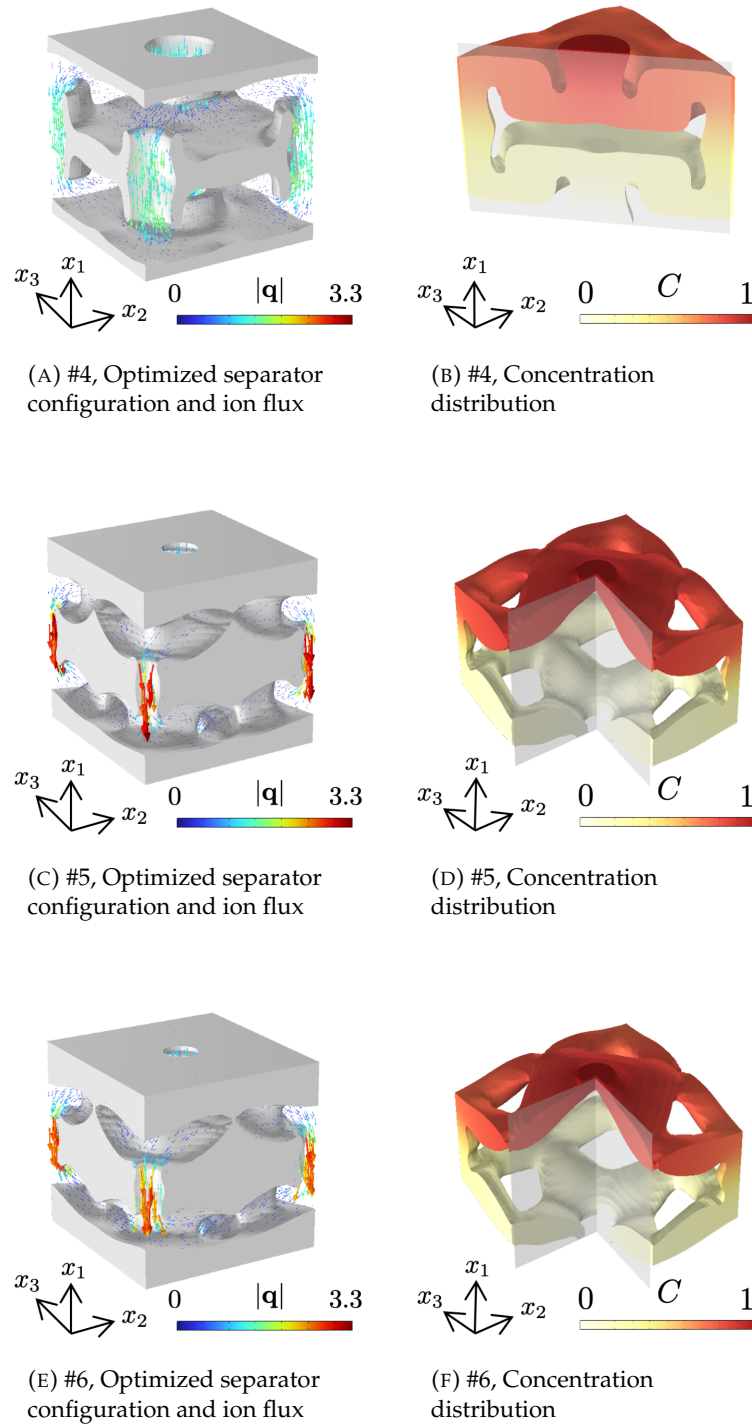


FIGURE 2.11: The optimized separator configurations, the ion flux, and the concentration distributions for #4, #5, and #6

Figure 2.11 shows the optimal separator structure and the region where the electrolyte exists, obtained by the optimization method executed under each condition. The boundary between the separator structure and the electrolyte is the isosurface of the design variable ρ at 0.5, and the red arrows in the figure showing the separator structure indicate the ion flux vector $\mathbf{q}$. As shown in Figs. 2.11a and b, the optimization result obtained without considering constraints shows that the electrolyte path exists in a curved manner within the fixed design domain with respect to the x_1 direction where the electrodes are located, and the tortuosity can be increased. Here, we compare the optimal structure obtained in three dimensions with the three-dimensional structure obtained by extruding the optimal structure obtained in two dimensions (Fig. 2.8b) in the x_3 direction. Evaluating the objective function of each structure, the values were -0.0670 and -0.0567, indicating that a better solution was obtained by optimizing in three dimensions. Also, comparing the cross-section of the optimal electrolyte path obtained in three dimensions at $x_2 + x_3 = 1$ (Fig. 2.11b) with the optimal structure obtained in two dimensions (Fig. 2.8b), it can be seen that they have similar shape characteristics. This indicates that by defining the design variables in a three-dimensional domain, the added dimension, the depth direction, is effectively utilized when curving the electrolyte path. However, this optimal result, as shown in Fig. 2.11a, has a separator structure that is not connected anywhere, making it impossible to actually fabricate.

To solve this problem, a stiffness constraint is considered as defined in Section 2.2.3. As shown in Fig. 2.11c, by considering the stiffness constraint, an optimal structure without floating regions could be obtained. This also shows that the introduction of a stiffness constraint is rational for a battery separator, which requires stiffness that can withstand dendrite growth. It should be noted that there is no guarantee that the stiffness constraint can completely eliminate floating solids.

Next, by comparing the results of #5 and #6, we consider the change in the optimal structure when the constraint value for ion conductivity $\tilde{N}_1$ is added. Comparing Figs. 2.11c and e, although the electrolyte paths are similar, the maximum value of the ion flux magnitude $|\mathbf{q}|$ is smaller in #6, which considers the constraint value $\tilde{N}_1$, indicating that the electrolyte path is thicker in #6. In other words, #6 has a structure that allows ions to pass more easily. However, comparing the objective function values, #5 shows a better result. This result is consistent with the fact that the structure that prevents dendrite growth and ion conductivity are in a trade-off relationship, and it supports the validity of the formulated optimization method. From these results, it has become clear that by using the proposed optimization method, a structure that effectively suppresses short circuits due to dendrite growth while maintaining stiffness under a specified ion conductivity constraint can be derived.

Finally, by comparing the computational costs of optimization based on automatic differentiation and optimization based on continuous adjoint sensitivity analysis, the necessity of continuous adjoint sensitivity analysis is demonstrated. The optimization using automatic differentiation was performed using the optimization module of COMSOL Multiphysics, while the optimization based on continuous adjoint sensitivity analysis was performed using COMSOL with MATLAB.

A Precision 7920 Rack equipped with an Intel(R) Xeon(R) Gold 6246R CPU (3.40 GHz) and 384 GB of memory was used to measure the computation time.

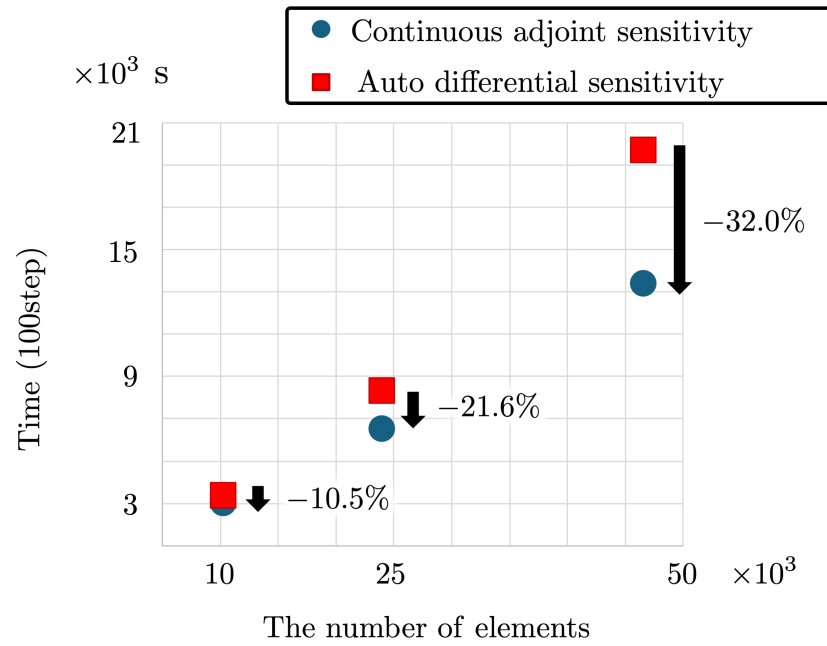


FIGURE 2.12: Comparison of computational costs with varying numbers of discretized elements in condition #4

Figure 2.12 shows a comparison of the computational costs under the optimization condition of #4. The horizontal axis represents the number of discrete elements, and the vertical axis represents the computation time for 100 optimization iterations. It was confirmed that as the number of elements increases, the difference in computational cost between optimization using continuous adjoint sensitivity analysis and optimization using automatic differentiation widens. Furthermore, this comparison was performed under the condition of #4, which does not involve the analysis of the displacement field, but in the conditions from #5 to #7, which consider stiffness as a constraint function, the number of degrees of freedom to be solved increases, so the difference in computational cost is expected to be even more significant.

From the above results, the importance of applying adjoint sensitivity analysis in the topology optimization of lithium-ion battery separators, which have diverse design requirements, was demonstrated.

2.4.4 Investigation for the objective function

In this section, the effectiveness of the objective function shown in Section 2.2.3 is discussed. In general, objective functions in fractional form are not actively adopted in optimization problems because they increase computational complexity. Looking at Section 2.2.3, it seems at first glance

that the same result could be obtained by optimizing only the numerator. Therefore, we define an evaluation function J_t using the numerator of Section 2.2.3 and compare the optimal results obtained when J_t and J are used as the respective objective functions.

$$J_t = - \int_D K(A\nabla C) \cdot (A\nabla C) dD. \quad (2.47)$$

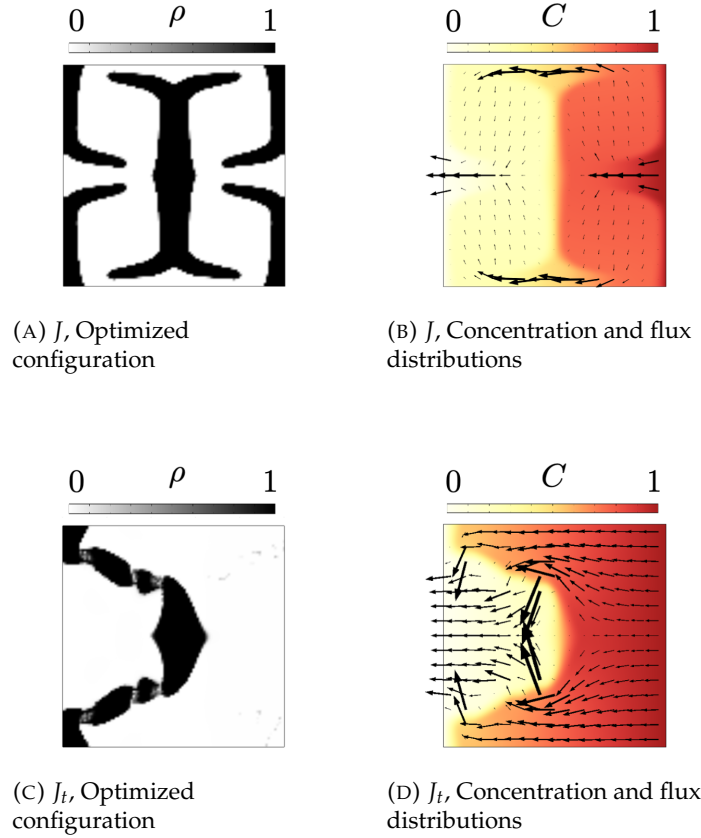


FIGURE 2.13: Optimized design variable configurations and distributions of concentration and flux (black arrows) for optimization problems with objective functions J and J_t

Figure 2.13 shows the optimal structures and the concentration distributions in those optimal structures obtained with the objective functions J_t and J , respectively. The initial structure used was the one shown in Fig. 2.8a. Comparing Figs. 2.13a and c, it can be confirmed that the obtained optimal structures are different. Comparing the ion flux distributions, it can be seen that the x_2 component of the ion flux is larger in Fig. 2.13d. Furthermore, evaluating J_t for each concentration distribution, the value was -6.83 for Fig. 2.13b and -11.256 for Fig. 2.13d.

From these results, it was shown that when only the numerator is used as the objective function, a locally narrow electrolyte path is obtained as a solution in an attempt to increase the magnitude of the ion flux vector in the direction of plane A. On the other hand, since the proposed objective

function evaluates the ratio of the magnitude of the gradients, the material is distributed so as to increase the ion flux in the direction of plane A throughout the unit cell domain, and it was confirmed to be an appropriate objective function for obtaining the desired structure. In addition, since the proposed objective function includes the magnitude of the concentration gradient in the denominator, it has the effect of reducing the concentration gradient at each point. This can prevent the formation of extremely narrow paths where the concentration gradient becomes excessively large, which may also be advantageous in terms of design and manufacturing.

2.5 Conclusions

A topology optimization method was developed for designing a battery separator that considers both stiffness and ion conductivity while suppressing short circuits caused by dendrite growth. The results are as follows:

- (1) The governing equations were formulated by focusing on the ion flux of impurity metals that cause dendrite growth, and an optimization problem was constructed to make dendrites grow in a plane perpendicular to the electrode direction. The sensitivity of the proposed objective function was derived in a continuous system using the adjoint variable method, and the effectiveness of the sensitivity derivation was demonstrated by comparing it with the sensitivity obtained in a discrete system in a numerical example.
- (2) An optimization algorithm based on the gradient method was developed for the formulated optimization problem.
- (3) Using the developed optimization algorithm, optimal structures obtained in two and three dimensions were presented, demonstrating that a structure that can suppress short circuits due to dendrite growth can be obtained by the proposed method.
- (4) It was shown that by considering the stiffness constraint, a manufacturable solution can be derived in which the separator structure is connected to the fixed boundaries. It was also shown that by considering the constraint on ion conductivity, an optimal solution can be derived from among the structures that exceed the desired ion conductivity.

It should be noted that to manufacture the optimal structure obtained by this method on a practical scale for lithium-ion batteries, a manufacturing technology capable of controlling a minimum feature size on the order of 10 nm is required.

Chapter 3

Topology optimization for all-solid-state-batteries using homogenization method[†]

CONTENTS

3.1	Introduction	51	3.4.1	Problem settings	65
3.2	Governing equations	54	3.4.2	Validation of proposed objective function	66
3.2.1	Analysis for all-solid-state-batteries	54	3.4.3	Verification of derived sensitivity .	69
3.2.2	Homogenization method	57	3.4.4	Numerical results	71
3.3	Topology optimization	59		Various C rates	71
3.3.1	Design variable settings	59		Effect on initial configurations . . .	72
3.3.2	Formulation of optimization problem	59		Effect on macrostructure configurations	74
3.3.3	Update scheme of design variable .	63		Considering two types of unit cells	75
3.3.4	Optimization algorithm	63	3.5	Conclusions	77
3.4	Numerical examples	64			

3.1 Introduction

With the increasing popularity of electric vehicles in recent times, the demand for more economical, higher-performance batteries as energy storage devices is rising. Although lithium-ion batteries (LIBs) have been widely used in portable devices or electronic vehicles due to their high energy density per unit weight, concerns towards the safety and the limitations of their energy capacities have evoked interest in improving the battery performance using different chemical compositions in battery development(Ouyang et al., 2019; Nagasubramanian and Fenton, 2013).

[†]The work in this chapter has been published in *Structural and Multidisciplinary Optimization*, DOI: 10.1007/s00158-024-03864-1.

Among them, All-Solid-State Batteries (ASSBs) exhibit excellent advantages over LIBs in terms of safety, energy density, and longevity, and bring a technological leap in the field of batteries(Ruan et al., 2022; Yuan et al., 2020; Zhang et al., 2017). As a main difference from LIBs, ASSBs employ solid electrolytes, which reduces the risk of leakage and combustion, and can have a higher energy density(Bates et al., 2022; Guo et al., 2022b). However, certain limitations of ASSBs also exist and hinder their practical applications. As the main issue, the ionic conductivity and electron mobility of ASSBs are inferior to those of LIBs since the contact surface area between the active material and solid electrolyte particles within composite electrodes may not be sufficiently equipped, which affects the charging or discharging rate(Tian and Qi, 2017). Another problem is that the ionic conductivity of solid electrolytes is lower than that of liquid electrolytes, increasing the internal resistance of the batteries(Zhang et al., 2023a).

Several researchers have proposed approaches to mitigate the abovementioned issues. These approaches are classified into three categories: i) material development, ii) improvement of process parameters, and iii) improvement of the design of material distributions. Regarding the development of materials including electrolytes and electrodes, solid electrolytes with better chemical/electrochemical compatibility with electrodes can improve the performance of ASSBs concerning long-term stability and higher ionic conductivity. Several research works revealed that coating the electrolytes with a different material increases the ionic conductivity and improves interface stability(Koç et al., 2022; Li et al., 2023). Other studies focused on developing materials that inhibit dendrite growth, which is critical for battery performance(Wei et al., 2023). It is well known that manufacturing batteries requires multiple processes, such as annealing and mechanical milling, and optimizing those manufacturing parameters can lead to higher manufacturing precision with battery performance. In the research work of (Tron et al., 2023; Zhang et al., 2023b), manufacturing parameters for specific battery manufacturing procedures have been optimized based on experimental results. As for the third category, inhomogeneously distributing active materials and electrolytes inside the battery can improve battery performance(Clausnitzer et al., 2023).

As a novel structural optimization method, Topology Optimization (TO) can provide the optimal material distribution that improves structural performance while satisfying certain constraints. Since the proposal of the pioneering research work by Bendsøe and Kikuchi, 1988, TO has been successfully applied to various physics problems, such as fluid problems(Borrvall and Petersson, 2003), reinforcement optimization, thermal-fluid system design(Matsumori et al., 2013), and acoustic meta-surface optimization(Noguchi et al., 2015).

Appropriate numerical analysis models, which provide an insight into the phenomena inside batteries, are necessary as guides to the design of structural topology inside the battery using TO. Many models have been proposed to simulate the electrochemical process within batteries. The Doyle-Fuller-Newman (DFN) model, also known as the Newman model, is the most widely

used as the mathematical framework(Doyle et al., 1993). It comprehensively accounts for lithium-ion transport in electrolytes, electron conduction in electrodes, and electrochemical reactions at electrode-electrolyte interfaces. In particular, the advantage of the DFN model lies in its possibility to accurately predict battery behavior under various operating conditions, making it a vital numerical model for battery design and analysis. Besides, several methods have been proposed to optimize the process parameters used in the DFN model to improve the accuracy and practicality of the analysis(Khalik et al., 2021; Jin et al., 2018). For instance, Danilov et al. have proposed the numerical analysis model for ASSBs with Li metal anode(Danilov et al., 2010). Moreover, for ASSBs with a graphite anode, Arunachalam et al. have focused on ionic conduction and lithium diffusion inside the battery are not isotropic and proposed the Full Homogenized Macroscale (FHM) model, in which the diffusion coefficient and the conductivity are defined using the homogenization method to account for anisotropy, based on the DFN model(Arunachalam et al., 2015). Compared to the DFN model, the FHM model presents better analysis results when considering high-temperature conditions(Arunachalam and Onori, 2019).

In the practical manufacturing process, designing internal material distribution design in batteries usually relies on the trial-and-error method, which consumes significant time and effort. To mitigate this issue, designing microstructures with enhanced structural performance using the TO technique is an effective method.

Many research achievements concerning various types of batteries have been made using TO. Iwai et al., 2011 proposed a TO method aimed at improving the efficiency of Solid Oxide Fuel Cells (SOFC), demonstrating its potential in applications. Yaji et al., 2018, Lin et al., 2022 and Roy et al., 2022 dealt with redox flow batteries, which have liquid electrolytes for the anode and cathode. They proposed a TO method to optimize the flow path inside redox batteries, further improving their efficiency. A TO method has also been proposed for the cooling plate to manage heat, which is problematic in batteries(Mo et al., 2021; Guo et al., 2022a). Pejman and Najafi have proposed the TO method for the structural battery composite(SBC) by assuming physics as the equivalent network(Pejman and Najafi, 2023). The SBC represents an innovative category of multifunctional materials, merging carbon fiber composites' load-supporting properties with a lithium-ion battery's energy storage function(Snyder et al., 2007; Snyder et al., 2008; Wong et al., 2007; Liu et al., 2009). Moreover, Alizadeh et al., 2023 have proposed the TO method for a porous reactor, which focused on the distribution of the volume fraction of the active and electrolyte materials.

Although some advancements have been achieved in the design of ASSBs, to the best of our knowledge, research concerning the structural optimization of ASSBs at the microscale level has not been found, and more detailed design guidelines have not yet been proposed. As a bridge to link microstructural configurations and macroscopic properties, the homogenization method is often used for the microstructure design assuming the periodic distribution of unit cells(Zhang et al., 2007; Yoo and Kikuchi, 2000; Nelli Silva et al., 1999; Feppon, 2024). Making use of this

characteristic, the combination of the homogenization method and the TO technique has the possibility to find the optimal material distribution for microstructures within ASSBs with macroscale performance.

In this paper, a new TO method for the design of the microstructures within ASSBs is constructed. Based on the reaction-diffusion equation-based level set method, the TO problem for ASSBs is formulated with design variables defined in a unit cell, that is assumed to align periodically in the composite region. First, a novel objective function is proposed, which can significantly facilitate the optimization process, based on the assumption that a uniform reaction within the composite region during the discharging process leads to more effective use of the active material. Compared to the commonly used objective function, it is shown that the proposed function has the same tendency as the original objective function and reduces the computational cost by one-half. The sensitivity is derived for the time-dependent and nonlinear problem using the Lagrange and continuous adjoint method. Subsequently, the effectiveness of the proposed TO method is studied by considering different C-rates, different initial microstructural guesses, and different macrostructures. Moreover, an optimization problem considering two types of unit cells in a composite anode region is implemented, and the results are presented. It should be mentioned here that although the application range of the proposed method is limited due to the simplification of the subject problem, to the best of our knowledge, this paper proposes the pioneering TO method for ASSBs based on the homogenization method.

The rest of the paper is organized as follows. Section 3.2 illustrates the mathematical formulation for the microstructure design of ASSBs, and the homogenization method is also introduced. Then, in Section 3.3, the TO problem based on the reaction-diffusion equation is defined, and the sensitivity derivation is presented. In Section 3.4, several numerical examples are provided to confirm the validity of the proposed approach. Finally, conclusions are provided in Section 3.5.

3.2 Governing equations

In this section, we explain the governing equation based on the analysis model for ASSBs using the homogenization method.

3.2.1 Analysis for all-solid-state-batteries

We construct the analysis model for ASSBs based on the DFN model(Doyle et al., 1993), which is widely used for battery analysis. Let us consider the 2D analysis domain in 2D, as Fig. 3.1 illustrates. ASSBs are composed of electrolyte Ω_l , composite anode Ω_n , and composite cathode Ω_p . The active material is distributed in the composite anode and cathode, whereas the electrolyte material is dispersed in the whole domain. The sum of the three regions is defined as Ω , with

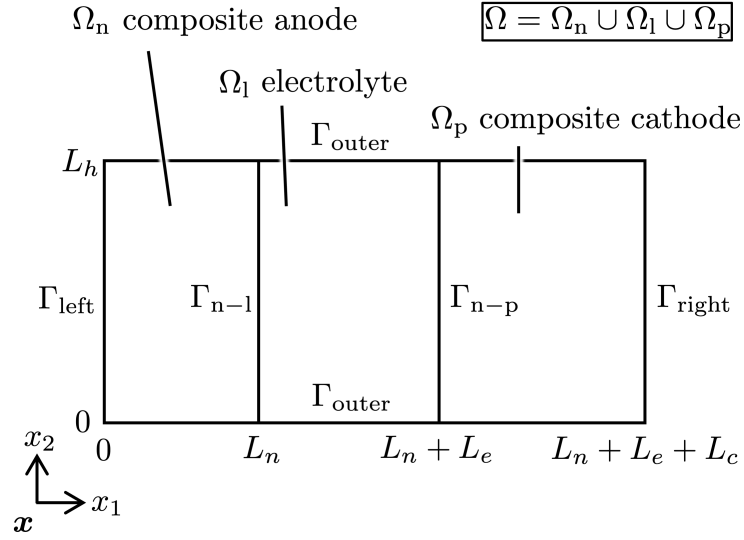


FIGURE 3.1: Schematics of the all-solid-state-battery

boundaries defined as Γ_{left} , Γ_{right} , and Γ_{outer} . The inner boundaries between the composite anode and the electrolyte, and between the composite cathode and the electrolyte, are defined as Γ_{n-1} and Γ_{n-p} , respectively. We can define the time variable t and spatial vector, as follows:

$$t \in \mathcal{T}[0, t_{\max}] \subseteq \mathbb{R}_{\geq 0} \quad (3.1)$$

$$\mathbf{x} = (x_1, x_2) \in \Omega \subseteq \mathbb{R}^2. \quad (3.2)$$

The inner variables to be considered in this model are concentration in the active material $c_{s,m}(\mathbf{x}, t; m = [n, p])$, electric potential in the active material $\phi_{s,m}(\mathbf{x}, t; m = [n, p])$, and electric potential in the electrolyte material $\phi_{e,m}(\mathbf{x}, t; m = [n, l, p])$. In the solid electrolyte system, the transference number is unity because the transported ion in the electrolyte is only Li^+ (KANNO, 2023). Then, the lithium transport equation in the composite electrodes can be written based on Fick's law as follows (Fuller and Harb, 2018):

$$\varepsilon_{s,m} \frac{\partial (c_{s,m})}{\partial t} = \nabla \cdot (D_{s,m}^{\text{eff}} \nabla c_{s,m}) - \frac{j_m^{\text{Li}}}{F} \quad \text{in } \mathcal{T} \times \Omega_m (m = n, p), \quad (3.3)$$

where ε_s , D_s^{eff} , and F are the volume fraction of active material, the effective lithium diffusion coefficient, and Faraday constant, respectively. j_m^{Li} is the charge transfer current density associated with the electrochemical reaction in the electrode and can be evaluated using the Butler-Volmer equation as follows (Fuller and Harb, 2018):

$$j_m^{\text{Li}} = a_s^{\text{eff}} i_{0,m} \left[\exp \left(\frac{0.5F}{RT} \eta_m \right) - \exp \left(-\frac{0.5F}{RT} \eta_m \right) \right], \quad (3.4)$$

where $a_s^{\text{eff}}[\text{m}^2/\text{m}^3]$ is the active surface area per electrode unit volume for charge-transfer reaction, $i_0[\text{A}/\text{m}^2]$ is the exchange current density, $R[\text{J}/(\text{mol} \cdot \text{K})]$ is the gas constant, and $T[\text{K}]$ is the temperature. $\eta[\text{V}]$ is the activation overpotential, as follows:

$$\eta_m = \phi_{s,m} - \phi_{e,m} - U_m(c_{s,m}), \quad (3.5)$$

where $U_m[\text{V}]$ is the equilibrium potential of each electrode. The charge conservation equation in the active material can be written as:

$$\nabla \cdot (\sigma_{s,m}^{\text{eff}} \nabla \phi_{s,m}) = j_m^{\text{Li}} \text{ in } \mathcal{T} \times \Omega_m(m = n, p), \quad (3.6)$$

where $\sigma_s^{\text{eff}}[\text{S}^{-1}\text{m}^{-1}]$ is the effective electron conductivity coefficient. Similarly, the charge conservation equation in the electrolyte can be written as:

$$\nabla \cdot (\kappa_m^{\text{eff}} \nabla \phi_{e,m}) = -j_m^{\text{Li}} \text{ in } \mathcal{T} \times \Omega_m(m = n, p), \quad (3.7)$$

where $\kappa^{\text{eff}}[\text{S}^{-1}\text{m}^{-1}]$ is the effective ion conductivity coefficient. It should be noted that the time derivative term in Eqs. (3.6) and (3.7) can be neglected because its time constant is smaller than that of the lithium transport equation(Doyle et al., 1993).

TABLE 3.1: Boundary conditions

Variable	Γ_{left}	Γ_{n-1}	Γ_{p-1}	Γ_{right}	Γ_{outer}
c_s	$D_{s,n}^{\text{eff}} \nabla c_{s,n} \cdot \mathbf{n} = 0$	$D_{s,n}^{\text{eff}} \nabla c_{s,n} \cdot \mathbf{n} = 0$	$D_{s,p}^{\text{eff}} \nabla c_{s,p} \cdot \mathbf{n} = 0$	$D_{s,p}^{\text{eff}} \nabla c_{s,p} \cdot \mathbf{n} = 0$	$D_s^{\text{eff}} \nabla c_s \cdot \mathbf{n} = 0$
ϕ_s	$\phi_{s,n} = 0$	$\sigma_{s,n}^{\text{eff}} \nabla \phi_{s,n} \cdot \mathbf{n} = 0$	$\sigma_{s,p}^{\text{eff}} \nabla \phi_{s,p} \cdot \mathbf{n} = 0$	$\sigma_{s,p}^{\text{eff}} \nabla \phi_{s,p} \cdot \mathbf{n} = -\frac{I}{A_g}$	$\sigma_s^{\text{eff}} \nabla \phi_s \cdot \mathbf{n} = 0$
ϕ_e	$\kappa_n^{\text{eff}} \nabla \phi_{e,n} \cdot \mathbf{n} = 0$	N/A	N/A	$\kappa_p^{\text{eff}} \nabla \phi_{e,p} \cdot \mathbf{n} = 0$	$\kappa^{\text{eff}} \nabla \phi_e \cdot \mathbf{n} = 0$

The boundary conditions for each variable are summarized in Table 3.1. The current value applied to the battery is $-I/A_g[\text{A}/\text{m}^2]$. Here, $A_g[\text{m}^2]$ is the area of boundary Γ_{right} and $I[\text{A}]$ is the magnitude of the current defined as:

$$I = \frac{Q_{\text{cell}}}{3600[\text{s}]} C_{\text{rate}}, \quad (3.8)$$

where C_{rate} is the parameter that determines the magnitude of the current. By the above definition, $1C_{\text{rate}}$ is the magnitude of the current that can fully discharge or charge the battery in 1 hour. On the other hand, $Q_{\text{cell}}[\text{C}]$ is the capacity of the battery calculated by the following equation:

$$Q_{\text{cell}} = c_{s,p,\text{max}} (\text{SOC}_{p,\text{max}} - \text{SOC}_{p,\text{min}}) \varepsilon_{s,p} V_p F, \quad (3.9)$$

where SOC is the ratio of the capacity at a given point to the capacity at full charge and $V_p[\text{m}^3]$ is the volume of the composite cathode.

The initial conditions for each concentration are defined as follows:

$$c_{s,p}(\mathbf{x}, t(0)) = c_{s,p,\max} \times \text{SOC}_{p,\min} \quad (3.10)$$

$$c_{s,n}(\mathbf{x}, t(0)) = c_{s,n,\max} \times \text{SOC}_{n,\max}. \quad (3.11)$$

In this paper, we set the fully charged battery as the initial condition.

The governing equations are solved until the terminal voltage falls below the threshold. Here, the terminal voltage is defined as the average of the solid electric potential $\phi_{s,p}$ on Γ_{right} since there can be situations where $\phi_{s,p}$ is not uniform on Γ_{right} . The termination criterion is set as follows:

$$\frac{\int_{\Gamma_{\text{right}}} \phi_{s,p} d\Gamma}{\int_{\Gamma_{\text{right}}} d\Gamma} \leq V_{\text{terminate}}, \quad (3.12)$$

where $V_{\text{terminate}}$ is a given parameter.

3.2.2 Homogenization method

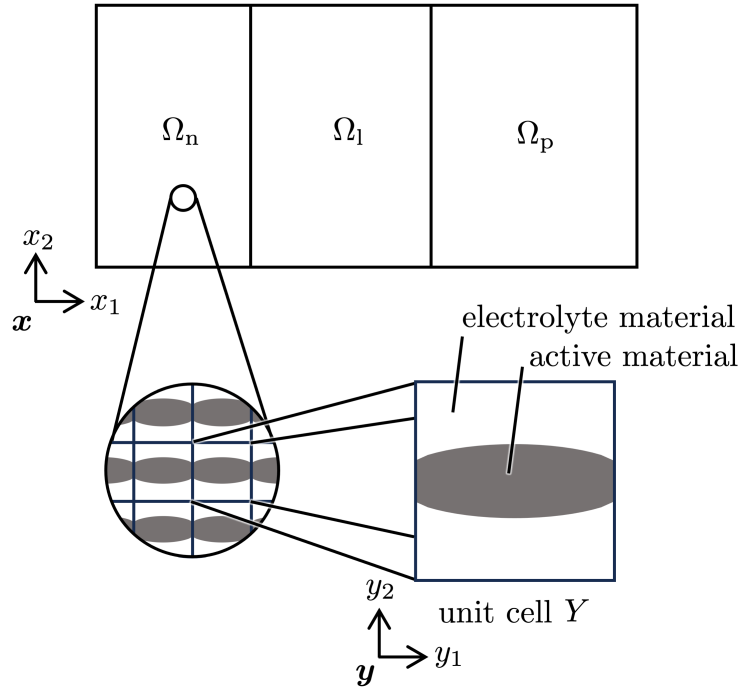


FIGURE 3.2: Schematics of the homogenization method

We explain a homogenization method based on the FHM model (Arunachalam et al., 2015). The homogenization method can derive macroscale material properties from the distribution of arbitrary active and electrolyte materials. In this study, it is considered that only the composite

anode has unit cells that are lined up periodically, as illustrated in Fig. 3.2. Therefore, the material property to be derived by the homogenization method is $D_{s,n}^{\text{eff}}$, $\sigma_{s,n}^{\text{eff}}$, and $\kappa_{s,n}^{\text{eff}}$, which become the second rank tensors instead of scalars to represent the anisotropic phenomenon. The unit cell Y , composed of the active and electrolyte material, is defined in the microscale $\mathbf{y} = (y_1, y_2) \in Y \subseteq \mathbb{R}^2$. Microscale $\mathbf{y}$ and macroscale $\mathbf{x}$ have the relation in the following equation:

$$\mathbf{y} = \frac{\mathbf{x}}{\omega_L}, \quad (3.13)$$

where ω_L is the ratio of the entire structure's representative length to the unit cell's representative length. Then, the Y -periodic admissible concentration field χ^D and the Y -periodic admissible electric potential fields, χ^σ and χ^κ , are defined within the unit cell. These characteristic variables exhibit periodicity in the y_1 and y_2 directions and are governed by the following equations:

$$\left\{ \begin{array}{ll} \nabla \cdot (D^{\text{unit}} \nabla \chi^D) = \nabla D^{\text{unit}} & \text{in } Y \quad (3.14a) \\ \chi^D : \text{periodic} & (3.14b) \\ \nabla \cdot (\sigma^{\text{unit}} \nabla \chi^\sigma) = \nabla \sigma^{\text{unit}} & \text{in } Y \quad (3.14c) \\ \chi^\sigma : \text{periodic} & (3.14d) \\ \nabla \cdot (\kappa^{\text{unit}} \nabla \chi^\kappa) = \nabla \kappa^{\text{unit}} & \text{in } Y \quad (3.14e) \\ \chi^\kappa : \text{periodic}, & (3.14f) \end{array} \right.$$

where D^{unit} , σ^{unit} , and κ^{unit} are the diffusion coefficient, the electronic conductivity, and the ionic conductivity, respectively. Based on the above variables, homogenized physical properties are obtained as in the following equation:

$$\left\{ \begin{array}{ll} D_{s,n}^{\text{eff}} = \frac{1}{|Y|} \int_Y D^{\text{unit}} \left\{ \tilde{I} - (\nabla \chi^D)^T \right\} dY & (3.15a) \\ \sigma_{s,n}^{\text{eff}} = \frac{1}{|Y|} \int_Y \sigma^{\text{unit}} \left\{ \tilde{I} - (\nabla \chi^\sigma)^T \right\} dY & (3.15b) \\ \kappa_{s,n}^{\text{eff}} = \frac{1}{|Y|} \int_Y \kappa^{\text{unit}} \left\{ \tilde{I} - (\nabla \chi^\kappa)^T \right\} dY & (3.15c) \\ \varepsilon_{s,n} = \frac{1}{|Y|} \int_Y \rho dY & (3.15d) \\ a_s^{\text{eff}} = \frac{1}{|Y|} \int_Y \rho a_s dY, & (3.15e) \end{array} \right.$$

where $\tilde{I}$ is the identity matrix, $a_s [\text{m}^2/\text{m}^3]$ is the active surface area per electrode unit volume for charge-transfer reaction, and the volume fraction of active material is calculated by integrating the design variable ρ within the unit cell, as shown in Eq. (3.15d).

3.3 Topology optimization

3.3.1 Design variable settings

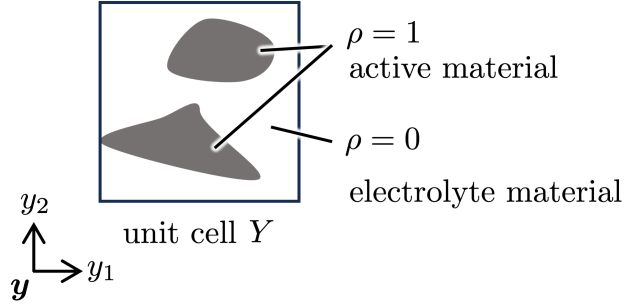


FIGURE 3.3: Design variable in the unit cell

In this study, we introduce the density method to represent the types of the material in the unit cell. Thus, we define the design variable $\rho(\mathbf{y})$ within the unit cell Y as Fig. 3.3 illustrates. $\rho = 1$ represents the region of active material, and $\rho = 0$ represents that of electrolyte material.

Active and electrolyte materials have different physical properties at each point within the unit cell Y according to the distribution of the design variable ρ . We utilize the Solid Isotropic Material with Penalization (SIMP) method (Bendsøe, 1989; Bendsøe and Sigmund, 1999) to continuously represent the physical properties D^{unit} , σ^{unit} , κ^{unit} , in the unit cell as follows:

$$\begin{cases} D^{\text{unit}} = D_{\text{act}}^{\text{unit}} \{ (1 - \omega_{\min}) \rho^3 + \omega_{\min} \} & (3.16a) \\ \sigma^{\text{unit}} = \sigma_{\text{act}}^{\text{unit}} \{ (1 - \omega_{\min}) \rho^3 + \omega_{\min} \} & (3.16b) \\ \kappa^{\text{unit}} = \kappa_{\text{ele}}^{\text{unit}} \{ (1 - \omega_{\min}) (1 - \rho)^3 + \omega_{\min} \}, & (3.16c) \end{cases}$$

where $D_{\text{act}}^{\text{unit}}$, $\sigma_{\text{act}}^{\text{unit}}$, and $\kappa_{\text{ele}}^{\text{unit}}$ are the diffusion coefficient in the active material, the electronic conductivity in the active material, and the ionic conductivity in the electrolyte material, respectively. The sufficiently small value $\omega_{\min}$ is considered in the other region to ensure stability in the numerical analysis.

3.3.2 Formulation of optimization problem

The optimization target to be considered in this study is the maximizing performance of the ASSBs. Figure 3.4 shows the example of charge-discharge curves, plotting the history of terminal voltage when the battery is charged and discharged. The area enclosed by these terminal voltage curves is the amount of energy lost during charging and discharging. We define this area as J_{ori} . We assume that a smaller area J_{ori} means less energy loss and hence a larger capacity can

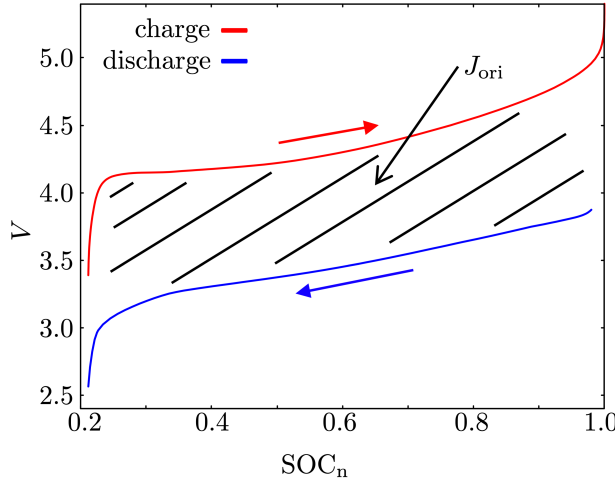


FIGURE 3.4: Charge-discharge curves

be extracted from the battery. In other words, minimizing this area could maximize the battery's efficiency. However, evaluating this area requires calculations for both charging and discharging processes, which leads to high computational costs in optimization problems that require iterative calculations. Therefore, we introduce an evaluation function that can be derived from the analysis of only the discharging process and has a distribution trend similar to the area enclosed by the curve. Here, we assume that a uniform reaction within the composite anode during the discharge process leads to more effective use of the active material in the composite anode, thereby reducing the energy loss within the battery. Therefore, we propose an objective function J_{pr} that focuses on the lithium concentration distribution, one variable that changes due to the electrode reaction, aiming to reduce the gradient of lithium concentration within the composite anode, as follows:

$$J_{pr} = \frac{\int_{\mathcal{T}} \int_{\Omega_n} (\nabla c_{s,n} \cdot \nabla c_{s,n}) d\Omega dt}{C_{pr} t_{max} V_n}, \quad (3.17)$$

where V_n is the area of composite anode Ω_n and C_{pr} is the parameter for normalizing the proposed objective function. The validity of the proposed objective function is discussed in Section 3.4.2. We thus formulate the optimization problem as:

$$\inf_{\rho} J = J_{pr} + J_{dw} \quad (3.18)$$

$$s.t. \quad \varepsilon_{s,n} - \varepsilon_{s,n,max} = 0 \quad (3.19)$$

$$0 \leq \rho \leq 1, \quad (3.20)$$

where $\varepsilon_{s,n,\max}$ is the target volume fraction of the active material in the unit cell, and J_{dw} is the double-well potential to accelerate the binarization of the design variable as follows:

$$J_{\text{dw}} = \int_Y \frac{C_{\text{dw}}}{4} \rho^2 (1 - \rho)^2 dY, \quad (3.21)$$

where C_{dw} is the parameter that determines the weight of the double-well potential.

Next, the sensitivity for the objective function is derived using the Lagrange and continuous adjoint methods. First, the Lagrangian is defined for the abovementioned optimization problem as follows:

$$\begin{aligned} \mathcal{L} = & J + \int_{\mathcal{T}} \int_{\Omega_n} a_1 \left(\varepsilon_{s,n} \frac{\partial (c_{s,n})}{\partial t} - \nabla \cdot (D_{s,n}^{\text{eff}} \nabla c_{s,n}) + \frac{j_n^{\text{Li}}}{F} \right) d\Omega dt \\ & + \int_{\mathcal{T}} \int_{\Omega_p} a_2 \left(\varepsilon_{s,p} \frac{\partial (c_{s,p})}{\partial t} - \nabla \cdot (D_{s,p}^{\text{eff}} \nabla c_{s,p}) + \frac{j_p^{\text{Li}}}{F} \right) d\Omega dt \\ & + \int_{\mathcal{T}} \int_{\Omega_n} a_3 \left(\nabla \cdot (\sigma_{s,n}^{\text{eff}} \nabla \phi_{s,n}) - j_n^{\text{Li}} \right) d\Omega dt \\ & + \int_{\mathcal{T}} \int_{\Omega_p} a_4 \left(\nabla \cdot (\sigma_{s,p}^{\text{eff}} \nabla \phi_{s,p}) - j_p^{\text{Li}} \right) d\Omega dt \\ & + \int_{\mathcal{T}} \int_{\Omega} a_5 \left(\nabla \cdot (\kappa_m^{\text{eff}} \nabla \phi_{e,m}) + j_m^{\text{Li}} \right) d\Omega dt \quad (m = n, l, p) \end{aligned} \quad (3.22)$$

where each $a_i = a_i(\mathbf{x}, t)$ is adjoint variable, respectively. The derivative of $\mathcal{L}$ by ρ is expressed by the following equation:

$$\begin{aligned} \frac{d\mathcal{L}}{d\rho} = & \frac{\partial \mathcal{L}}{\partial \rho} + \frac{\partial \mathcal{L}}{\partial c_{s,n}} \frac{\partial c_{s,n}}{\partial \rho} + \frac{\partial \mathcal{L}}{\partial c_{s,p}} \frac{\partial c_{s,p}}{\partial \rho} \\ & + \frac{\partial \mathcal{L}}{\partial \phi_{s,n}} \frac{\partial \phi_{s,n}}{\partial \rho} + \frac{\partial \mathcal{L}}{\partial \phi_{s,p}} \frac{\partial \phi_{s,p}}{\partial \rho} + \frac{\partial \mathcal{L}}{\partial \phi_e} \frac{\partial \phi_e}{\partial \rho}, \end{aligned} \quad (3.23)$$

Given the difficulty in computing $\partial c_{s,n} / \partial \rho$, we aim to determine the distribution of a_1 for which the condition $\partial \mathcal{L} / \partial c_{s,n} = 0$ is satisfied. Thus, the equation for the adjoint variable a_1 can be derived as:

$$\left\{ \begin{array}{l} -\varepsilon_{s,n} \frac{\partial a_1}{\partial t} - \nabla \cdot (D_{s,n}^{\text{eff}} \nabla a_1) + A \left(-M \frac{\partial U_n(c_{s,n})}{\partial c_{s,n}} \right) \\ \quad - \frac{2}{C_{\text{pr}} t_{\text{max}} V_n} \nabla \cdot (\nabla c_{s,n}) = 0 \quad \text{in } \mathcal{T} \times \Omega_n \end{array} \right. \quad (3.24a)$$

$$\left\{ \begin{array}{l} D_{s,n}^{\text{eff}} \nabla a_1 \cdot \mathbf{n} = -\frac{2}{C_{\text{pr}} t_{\text{max}} V_n} \nabla c_{s,n} \cdot \mathbf{n} \quad \text{on } \mathcal{T} \times \partial \Omega_n \end{array} \right. \quad (3.24b)$$

$$\left\{ \begin{array}{l} a_1(t(t_{\text{max}})) = 0 \quad \text{in } \Omega_n \end{array} \right. \quad (3.24c)$$

where $\partial\Omega_n$ is the boundary of all the enclosed Ω_n , and A and M are the variables that are defined as follows:

$$A = \begin{cases} \frac{a_1}{F} - a_3 + a_5 & \text{in } \Omega_n \\ 0 & \text{in } \Omega_l \\ \frac{a_2}{F} - a_4 + a_5 & \text{in } \Omega_p, \end{cases} \quad (3.25)$$

$$M = \frac{\partial j^{\text{Li}}}{\partial \eta} = a_s^{\text{eff}} i_{0,m} \left[\frac{0.5F}{RT} \exp\left(\frac{0.5F}{RT} \eta\right) + \frac{0.5F}{RT} \exp\left(-\frac{0.5F}{RT} \eta\right) \right], \quad (3.26)$$

A detailed derivation of Eq. (3.24) is given in Appendix A.2. Similarly, under the conditions $\partial\mathcal{L}/\partial c_{s,p} = 0$, $\partial\mathcal{L}/\partial\phi_{s,n} = 0$, $\partial\mathcal{L}/\partial\phi_{s,p} = 0$, and $\partial\mathcal{L}/\partial\phi_e = 0$, the equations for the adjoint variables a_2 , a_3 , a_4 , and a_5 can be derived as:

$$\begin{cases} -\varepsilon_{s,p} \frac{\partial a_2}{\partial t} - \nabla \cdot (D_{s,p}^{\text{eff}} \nabla a_2) & \end{cases} \quad (3.27a)$$

$$\begin{cases} + A \left(-M \frac{\partial U_n(c_{s,p})}{\partial c_{s,p}} \right) = 0 & \text{in } \mathcal{T} \times \Omega_p \end{cases} \quad (3.27b)$$

$$\begin{cases} D_{s,p}^{\text{eff}} \nabla a_2 \cdot \mathbf{n} = 0 & \text{on } \mathcal{T} \times \partial\Omega_p \end{cases} \quad (3.27c)$$

$$\begin{cases} a_2(t(t_{\text{max}})) = 0 & \text{in } \Omega_p, \end{cases} \quad (3.27d)$$

$$\begin{cases} \nabla \cdot (\sigma_{s,n}^{\text{eff}} \nabla a_3) + AM = 0 & \text{in } \mathcal{T} \times \Omega_n \end{cases} \quad (3.28a)$$

$$\begin{cases} a_3 = 0 & \text{on } \mathcal{T} \times \Gamma_{\text{left}} \end{cases} \quad (3.28b)$$

$$\begin{cases} \sigma_{s,n}^{\text{eff}} \nabla a_3 \cdot \mathbf{n} = 0 & \text{on } \mathcal{T} \times \partial\Omega_n \setminus \Gamma_{\text{left}}, \end{cases} \quad (3.28c)$$

$$\begin{cases} \nabla \cdot (\sigma_{s,p}^{\text{eff}} \nabla a_4) + AM = 0 & \text{in } \mathcal{T} \times \Omega_p \end{cases} \quad (3.29a)$$

$$\begin{cases} \sigma_{s,p}^{\text{eff}} \nabla a_4 \cdot \mathbf{n} = 0 & \text{on } \mathcal{T} \times \partial\Omega_p, \end{cases} \quad (3.29b)$$

$$\begin{cases} \nabla \cdot (\kappa_m^{\text{eff}} \nabla a_5) - AM = 0 & \text{in } \mathcal{T} \times \Omega_m \end{cases} \quad (3.30a)$$

$$\begin{cases} \kappa_m^{\text{eff}} \nabla a_5 \cdot \mathbf{n} = 0 & \text{on } \mathcal{T} \times \partial\Omega, \end{cases} \quad (3.30b)$$

where $\partial\Omega_p$ and $\partial\Omega$ are the boundaries of all the enclosed Ω_p and Ω , respectively. Then, sensitivity can be derived as:

$$\begin{aligned}
\frac{d\mathcal{L}}{d\rho} &= \frac{\partial \mathcal{L}}{\partial \rho} \\
&= - \int_{\Omega_n} \frac{\varepsilon_{s,n}}{\partial \rho} a_1 c_{s,n} \Big|_{t=0} d\Omega \\
&\quad + \int_{\mathcal{T}} \int_{\Omega_n} \left(-\frac{\partial \varepsilon_{s,n}}{\partial \rho} c_{s,n} \frac{\partial a_1}{\partial t} + \frac{\partial D_{s,n}^{\text{eff}}}{\partial \rho} \nabla a_1 \cdot c_{s,n} - \frac{\partial \sigma_{s,n}^{\text{eff}}}{\partial \rho} \nabla a_3 \cdot \phi_{s,n} \right. \\
&\quad \quad \left. - \frac{\partial \kappa_n^{\text{eff}}}{\partial \rho} \nabla a_5 \cdot \phi_{e,n} + \left(\frac{a_1}{F} - a_3 + a_5 \right) \frac{\partial j^{\text{Li}}}{\partial \rho} \right) d\Omega dt \\
&\quad + \int_Y C_{\text{dw}} \rho (1 - \rho) \left(\frac{1}{2} - \rho \right) dY \\
&:= \int_Y J' dY,
\end{aligned} \tag{3.31}$$

where J' is the sensitivity function defined within the unit cell Y . The derived sensitivity is validated by comparing it with the finite difference in Section 3.4.3.

3.3.3 Update scheme of design variable

This paper introduces the reaction-diffusion equation-based topology optimization method, proposed by Choi et al., 2011, to obtain the optimum point in the feasible region. The design variable ρ is considered as the function depends on the fictitious time t_{rde} and is updated by the reaction-diffusion equation as follows:

$$\begin{cases} \frac{\partial \rho}{\partial t_{\text{rde}}} = - (C_{\text{diff}} J' - \tau \nabla^2 \rho) & \text{in } Y \\ \rho : \text{periodic} & \text{on } \partial Y, \end{cases} \tag{3.32a}$$

$$\tag{3.32b}$$

where C_{diff} is the normalization parameter for sensitivity J' , and τ is the regularization parameter to guarantee the smoothness of the design variable during the optimization process. The periodic boundary condition is applied on the outer boundary of the unit cell to ensure the connectivity for each material region.

3.3.4 Optimization algorithm

In this section, the optimization algorithm we introduced is described. Figure 3.5 summarizes the schematic of the optimization algorithm. First, the design variable in the unit cell is initialized by giving a specific distribution. Then, the equations for the homogenization method are solved based on the design variable to obtain the Y -periodic admissible concentration and electric potential fields. Subsequently, the homogenized material properties D^H , σ^H , and κ^H are calculated

using the equations. Then, the time-dependent governing equations for ASSBs are solved until Eq. (3.12) is satisfied. The objective function J and constraint function are calculated using the obtained variables. Subsequently, the convergence of the optimization problem is evaluated by the following equation:

$$\left\| \frac{J_{i+1} - J_i}{J_i} \right\| \leq \omega, \quad (3.33)$$

where ω is the threshold for determining convergence. If Eq. (3.33) is satisfied, the optimization is terminated; if not, the time-dependent adjoint equations are solved to obtain the sensitivity. Then, the design variable is updated by the sensitivity using the reaction-diffusion equation, Eq. (3.32a), and the process returns to the first step.

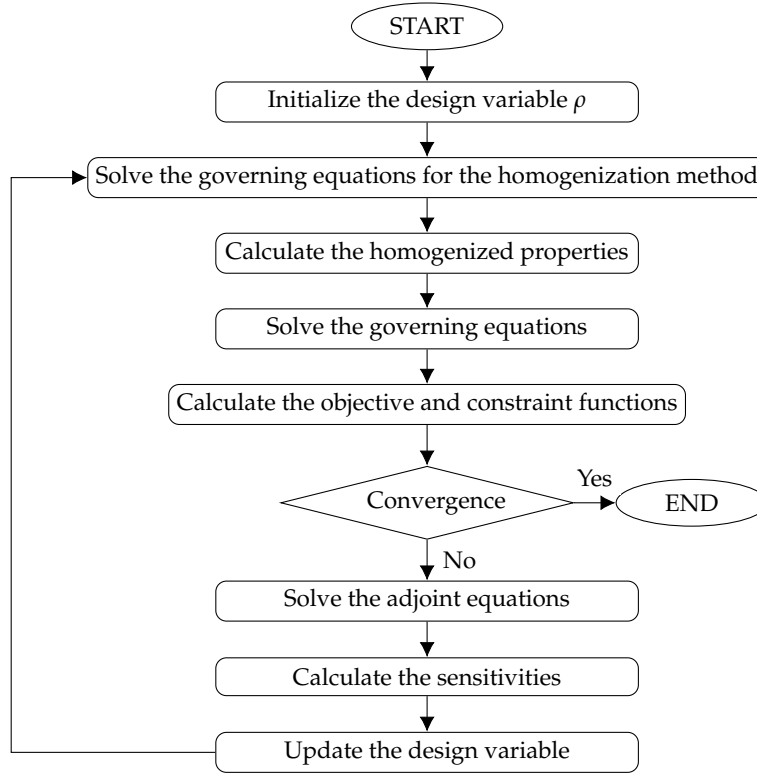


FIGURE 3.5: Optimization flow chart.

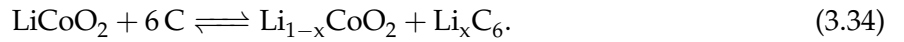
3.4 Numerical examples

This section presents numerical examples of the optimization problems defined earlier, demonstrating the effectiveness of our research. Sect. 3.4.1 outlines the problem settings. Following this, Section 3.4.2 provides a comparison between the proposed and original objective functions. Finally, Sect. 3.4.4 presents the results obtained using the proposed optimization method under

various conditions. We conduct the numerical examples using COMSOL Multiphysics Simulations with Matlab®. Specifically, the finite element analysis is conducted in COMSOL, and the optimization algorithm is conducted in Matlab.

3.4.1 Problem settings

This study assumes that the cathode is lithium cobalt oxide and the anode is graphite carbon. The equilibrium potentials U_n and U_p are set based on the material data sourced from COMSOL Multiphysics, 1998. The intercalation and deintercalation reaction that occurs in the battery is expressed by the following chemical equation:



The left-to-right side is the discharge reaction, which can extract energy, and the right-to-left side is the charge reaction, which can store energy.

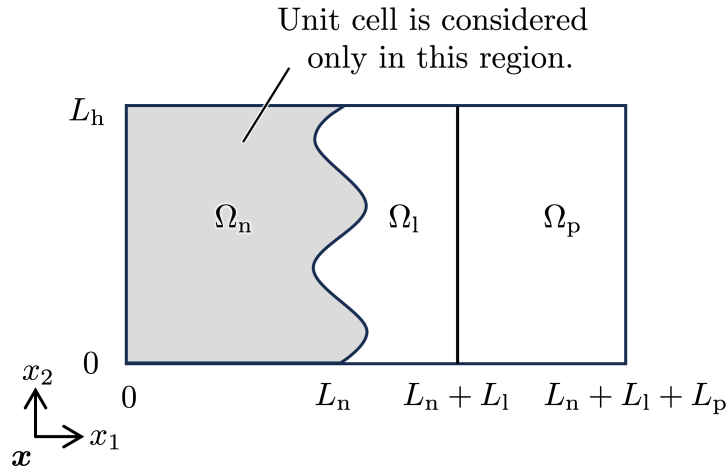


FIGURE 3.6: Schematics of the macro scale structure, type B

The values of the parameters used are summarized in Table 3.2, where C_{dw} is varied stepwise during the optimization time step to ensure better convergence. The temperature is assumed to be constant in this study. We set three macrostructure configurations as types A, B, and C, where the following three equations represent the interface between the anode and the electrolyte.

$$\left\{ \begin{array}{ll} \text{type A :} & x_1 = L_n \end{array} \right. \quad (3.35a)$$

$$\left\{ \begin{array}{ll} \text{type B :} & x_1 = Q \sin \left(\frac{2\pi}{L_h} n x_2 \right) + L_n \end{array} \right. \quad (3.35b)$$

$$\left\{ \begin{array}{ll} \text{type C :} & x_1 = \frac{2}{5} \left(x_2 - \frac{L_h}{2} \right) + L_n, \end{array} \right. \quad (3.35c)$$

where Q and n are the amplitude and frequency of the sin function, respectively. As an example, the macrostructure of type B with $Q = 10[\mu\text{m}]$, $n = 2$ is shown in Fig. 3.6. The unit cell Y is considered only in the composite anode Ω_n in this study.

The macrostructure is discretized with triangle unstructured mesh, as Fig. 3.7a shows. The variables in the macrostructure are defined in the finite element space discretized with quadratic shape functions. On the other hand, the unit cell is discretized with quadrangle structured mesh, as Fig. 3.7b shows. The size of one element is set to 0.02. The variables in the unit cell are defined in the finite element space discretized with linear shape functions.

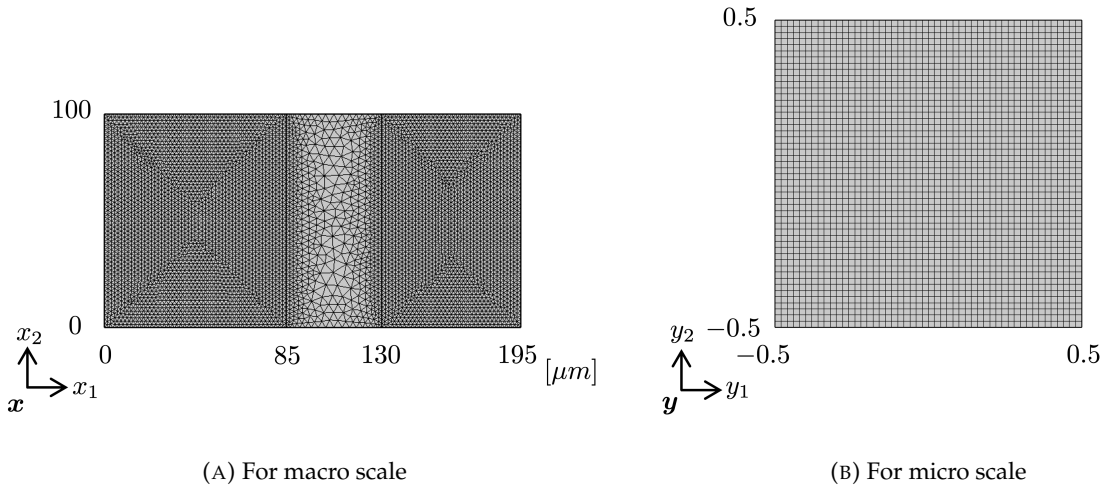


FIGURE 3.7: Meshes for each scale geometry

3.4.2 Validation of proposed objective function

In this section, we confirm the validity of the objective function proposed in Section 3.3.2. As mentioned in Section 3.3.2, the area enclosed by the charge-discharge curve in Fig. 3.4 is used to assess the performance of a battery. We compare the proposed objective function J_{pr} with the original objective function J_{ori} when varying the macrostructure and microstructure configurations. The interface between the anode and the electrolyte is expressed by type B, Eq. (3.35b), and we alter the macrostructure configuration by changing the values of Q and n to $[8, 10, 12, 14]$ and $[1, 2, 3, 4, 5]$, respectively. Calculations are performed under the conditions of 0.5C, 1C, and 2C, and the results obtained are normalized and compared, as shown in Fig. 3.8. Here, the horizontal axis of the plots represents the index numbers corresponding to the sequential combinations of Q and n values. Q values are varied in the order of $[8, 10, 12, 14]$, and for each Q , n values are varied in the order of $[1, 2, 3, 4, 5]$. Each combination of Q and n is assigned a unique index number, plotted sequentially along the horizontal axis. In these results, although there are some differences at 2C,

TABLE 3.2: Parameter settings

Parameter	Value	Unit
L_n	85.0	m
L_l	45.0	m
L_p	65.0	m
L_h	100	m
$D_{s,p}^{\text{eff}}$	5.00×10^{-13}	m ² /s
$\sigma_{s,p}^{\text{eff}}$	3.30×10^{-2}	S/m
κ_l^{eff}	2.00×10^{-2}	S/m
κ_p^{eff}	3.30×10^{-2}	S/m
C_{dw}	[2.0, 5.0, 7.0, 9.0]	–
C_{diff}	1.0	–
C_{pr}	1×10^{15}	–
τ	5×10^{-3}	–
$\varepsilon_{s,p}$	0.44	–
$\varepsilon_{s,n,\text{max}}$	0.3	–
ω_{min}	1.0×10^{-3}	–
V_p	6.5×10^{-9}	m ²
F	96485.3	C/mol
R	8.3143	J/(mol · K)
$\text{SOC}_{p,\text{max}}$	1	–
$\text{SOC}_{p,\text{min}}$	0.43	–
$\text{SOC}_{n,\text{max}}$	0.98	–
$\text{SOC}_{n,\text{min}}$	0	–
$c_{s,p,\text{max}}$	56250	mol/m ³
$c_{s,n,\text{max}}$	31507	mol/m ³
a_s	1×10^6	m ² /m ³
$i_{0,p}$	26	A/m ²
$i_{0,n}$	36	A/m ²
T	298	K
$V_{\text{terminate}}$	2.7	V
ω	1.0×10^{-3}	–
$D_{\text{act}}^{\text{unit}}$	1.45×10^{-13}	m ² /s
$\sigma_{\text{act}}^{\text{unit}}$	100	S/m
$\kappa_{\text{ele}}^{\text{unit}}$	2.00×10^{-2}	S/m

the distribution of the proposed objective function generally aligns with that of the original objective function. Similarly, comparisons are made by altering the angle θ between the y_1 -axis and the

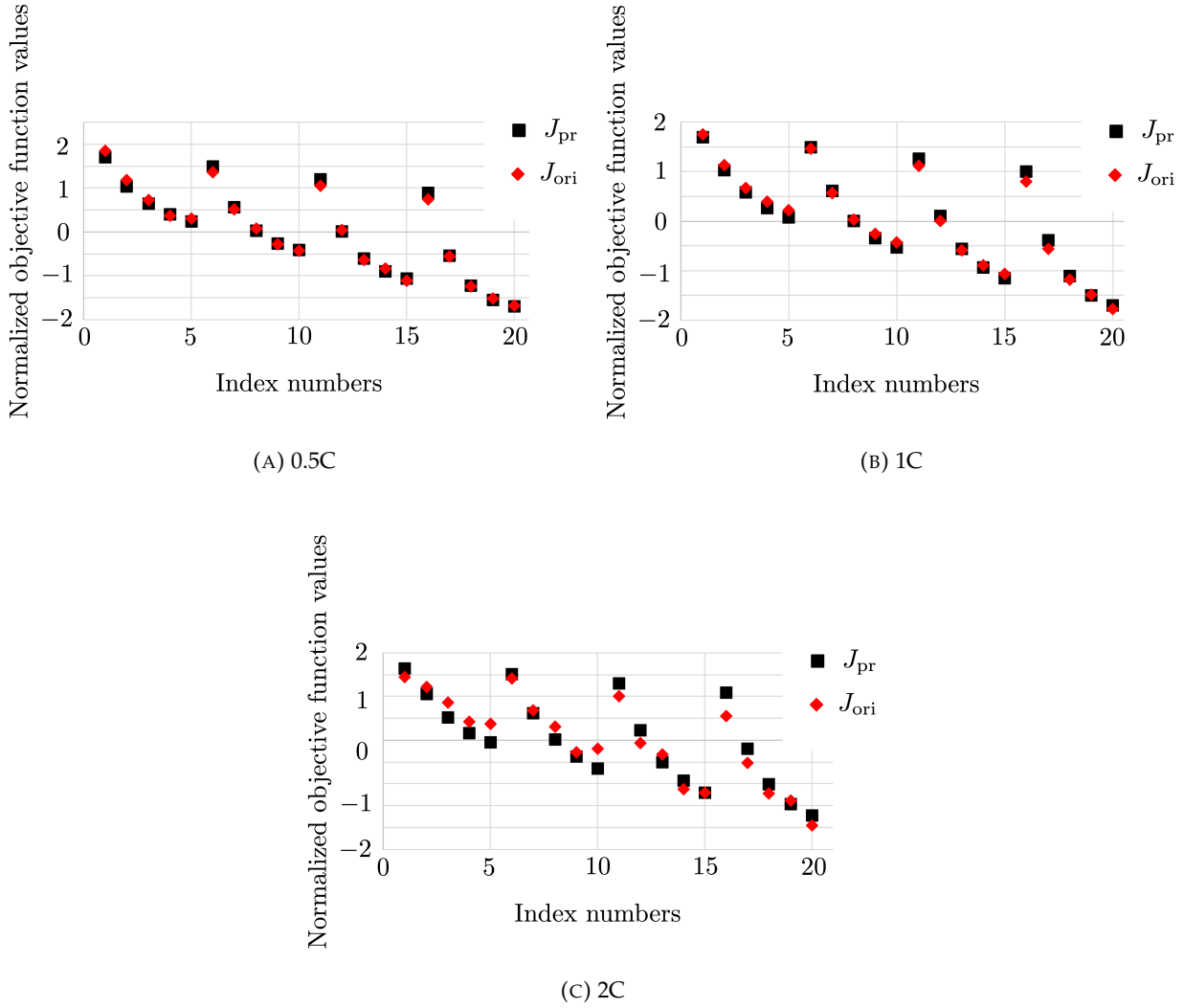


FIGURE 3.8: Plots for the normalized proposed and original objective functions for 0.5C, 1C and 2C across varying macrostructure configurations with Q [8, 10, 12, 14] and n [1, 2, 3, 4, 5]

active material distribution in the microstructure, as shown in Fig. 3.9. We vary the value of $\tan \theta$ across ten values [0, 0.2, 0.5, 0.8, 1, 1.25, 2, 5, 100, 200], while keeping the area of the active material within the unit cell constant. Calculations are conducted under the 1C condition, and the results of the normalized objective function values are presented in Fig. 3.10. Here, the horizontal axis of the plot represents the index numbers assigned to the varying values of $\tan \theta$, ranging from 1 to 10, corresponding to the order of the values [0, 0.2, 0.5, 0.8, 1, 1.25, 2, 5, 100, 200]. It is observed that even when altering the microstructure, the distribution of the proposed objective function generally matches that of the original objective function. Moreover, the computational cost comparison for calculating J_{ori} and J_{pr} is conducted. As mentioned in Sect. 4, J_{ori} needs the numerical analysis

for both charge and discharge processes, while J_{pr} needs the numerical analysis only for either the charge or discharge process. It takes 265s for J_{ori} and 68s for J_{pr} based on a desktop equipped with Intel® Xeon® Gold 6246R CPU with 32 cores, a clock frequency of 3.40GHz, and 256GB of memory, i.e., using J_{pr} has 74% calculation cost reduction from using J_{ori} . These comparisons show that the proposed objective function effectively captures the original objective function's distribution characteristics and is valid as a substitute for the objective function.

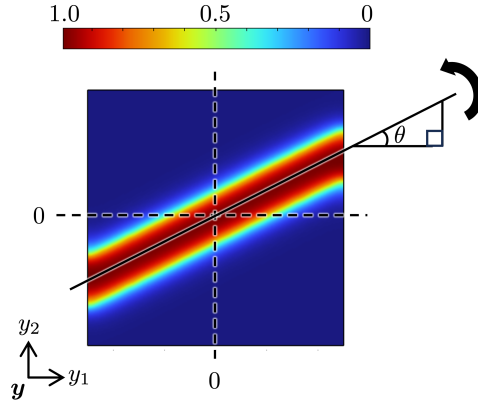


FIGURE 3.9: Material distributions within the unit cell when the gradient of the distributed active material is 0.5

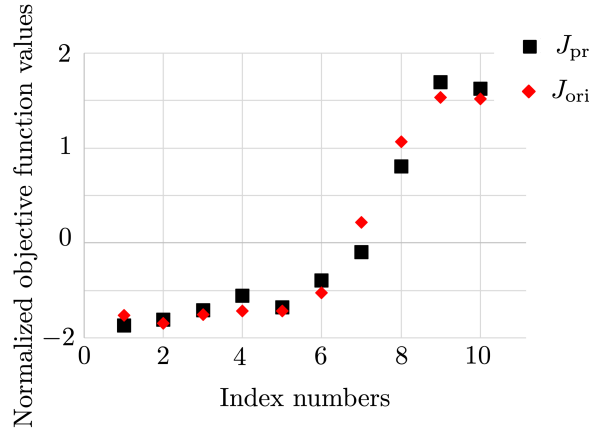


FIGURE 3.10: Plots for the normalized proposed and original objective functions for 1C when the angle θ between the y_1 -axis and the active material distribution in the unit cell is changed

3.4.3 Verification of derived sensitivity

In this section, we compare the derived sensitivity J' in Eq. (3.31) with the finite difference to confirm the validity of the derived sensitivity. The finite difference $\tilde{J}_{pr}$ can be defined as the following equation:

$$\tilde{J}_{pr}(\rho, \delta\rho) = \frac{J(\rho + \delta\rho) - J(\rho - \delta\rho)}{\int_Y(\rho + \delta\rho) dY - \int_Y(\rho - \delta\rho) dY}, \quad (3.36)$$

where $\delta\rho$ is the difference amount of the design variable ρ with a created small hole of radius. In this numerical example, $\delta\rho$ and the small hole radius are set to 0.1 and 0.02, respectively. $\delta\rho$ is set to 0.1 because it had the smallest maximum relative error among the sets $[0.05, 0.1, 0.2, 0.3]$. As Fig. 3.11 illustrates, we compare J' and $\tilde{J}_{\text{pr}}$ along cut lines (1) $y_2 = 0$ and (2) $y_1 = 0$ based on the set design variable distribution ρ . Here, C_{dw} is set to 0 because J_{dw} is a function depending only on ρ and the derivative of J_{dw} for ρ is obvious.

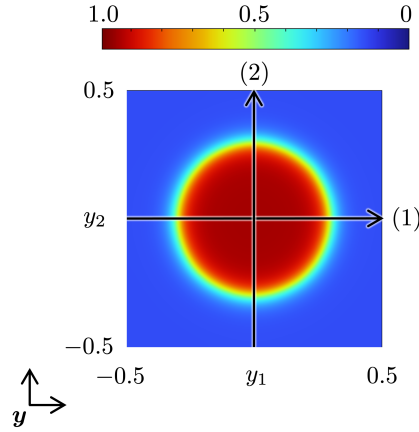


FIGURE 3.11: Design variable distribution ρ and the cut lines for the comparison of the derived sensitivity and the numerical difference

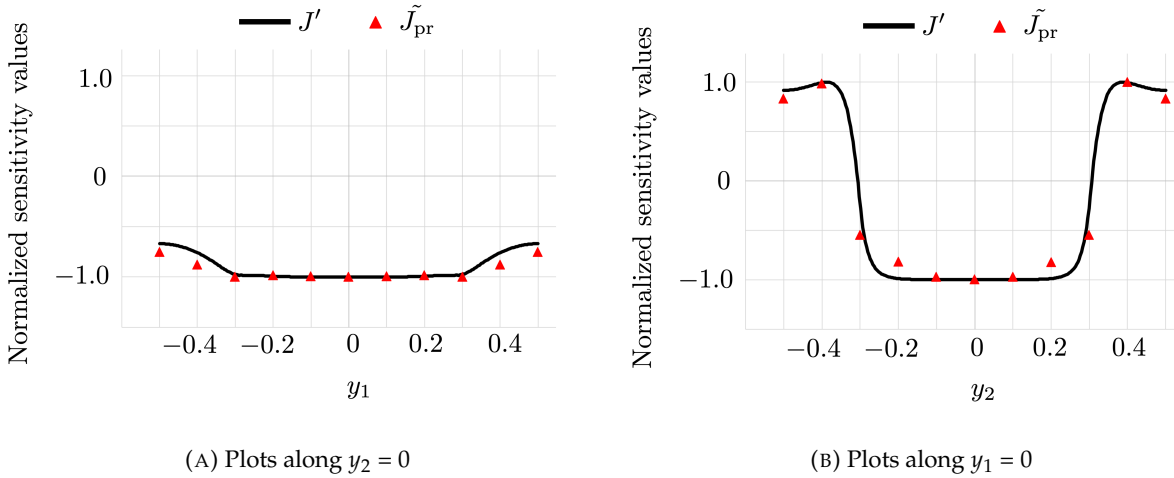


FIGURE 3.12: Plots for the normalized sensitivity J' and numerical difference $\tilde{J}_{\text{pr}}$ along the cut lines

Figure 3.12 plots the sensitivity J' and numerical difference $\tilde{J}_{\text{pr}}$ normalized by each maximum and minimum value. Assuming the numerical difference is correct, the maximum relative error of adjoint sensitivity is 49.4% at $(y_1, y_2) = (0, -0.3)$. This position is where ρ changes, as Fig. 3.11 shows, and where the distribution of sensitivity changes significantly. This resulted in a large numerical error. Though differences exist, the plots show that the deduced sensitivity using the Lagrange and continuous adjoint methods can follow the finite difference tendencies.

3.4.4 Numerical results

Various C rates

In this example, three different C rates are considered, i.e., 0.5C, 1C, and 2C. The corresponding optimization results are given in Fig. 3.13. Despite certain gray elements, the active material in the optimized results among three cases are distributed in the horizontal direction. From the view of the authors, such material distribution is reasonable considering the chemical reaction within the ASSBs. With the horizontal layout of the active material, the diffusion coefficient in the horizontal direction becomes more prominent than the case of vertical distribution. In other words, lithium ions are transported to the electrode in a higher efficiency when the electrolyte is distributed horizontally with electrons are transferred to the electrolyte phase much closer, which contributes to a more uniform electrochemical reaction within the battery.

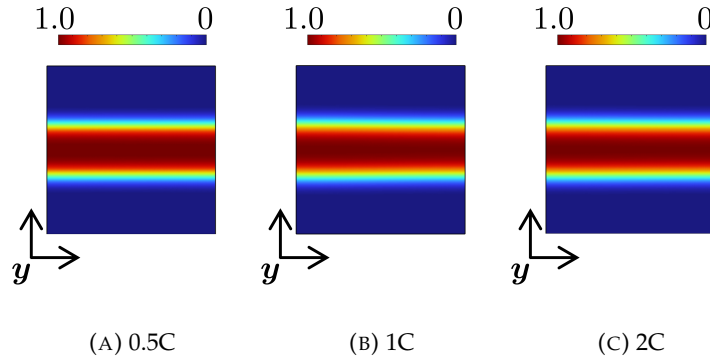


FIGURE 3.13: The obtained results for 0.5C, 1C and 2C

Subsequently, we conduct a cross-checking examination to discuss the characteristics of the proposed method. We perform the original objective function J_{ori} under 0.5C, 1C, and 2C condition for the results obtained by the proposed optimization method under 0.5C, 1C, and 2C. The results are summarized in Table 3.3. The optimal solution obtained at the 0.5C condition performs well under both 0.5C and 1C conditions. Meanwhile, the optimal solutions identified for the 1C and 2C conditions show favorable outcomes, specifically for the 2C condition. According to the author's view, as mentioned in Section 3.4.2, the reason the optimal solution at the 0.5C condition also performs well under the 1C condition is attributed to the fact that slower charging and discharging rates facilitate a more precise capture of changes in internal variables, thereby leading to a more accurate optimization outcome. It should be noted that the outcome of this result does not imply that optimization conducted at 0.5C will invariably yield superior results under all possible conditions.

TABLE 3.3: Original objective function values J_{ori} analyzed at 0.5C, 1C, and 2C for structures obtained at 0.5C, 1C, and 2C in the optimization, respectively

optimization \ calculation	0.5C	1C	2C
	0.5C	0.515	0.847
1C	0.516	0.850	0.90
2C	0.516	0.850	0.90

Effect on initial configurations

In this subsection, the influence of the initial microstructural guess on optimization results is examined. As shown in Fig. 3.14, three different initial microstructural guesses are considered, with the active material distributed as (a) a vertically elongated ellipse, (b) a horizontally elongated ellipse, and (c) four circles. The corresponding optimization results are presented in Fig. 3.15. In all cases, as in the results obtained in Sect. 3.4.4, the active material and electrolyte are distributed horizontally. The results starting from a single ellipse, (a) and (b), showed that the active material distributed in one large mass, whereas starting from four circles resulted in the active material dividing into two parts. These results indicate that the optimal structure depends on the initial number of active material pieces, which stems from the limitations in the formulation of the homogenization method. As shown in equation Eq. (3.13), ω_L relates the microscale y and macroscale x in the homogenization method. However, in the derivation of Eqs. (3.14) and (3.15), ω_L is assumed to be sufficiently small, and terms of order higher than ω_L are ignored. As a result, ω_L is not included in Eqs. (3.14) and (3.15). Comparing Figs. 3.15b and c, if the solution in Fig. 3.15c is divided into four parts, it becomes the same structure as in Fig. 3.15b. In other words, the macroscopic representation of microstructures shown in Fig. 3.15b with $\omega_L/2$ will be the same as the one given in Fig. 3.15c with ω_L .

To check the convergence performance of the proposed method, Fig. 3.16 shows the iteration history of the objective and constraint function values using the initial guess (a) and (b). For reasonable comparison, the value of C_{dw} is changed at the exact timing in both cases. Generally, the optimization using two different initial microstructures presents similar iteration performance, and in this result, the objective function's histories become stable around the 250th step. Besides, it can be observed that the objective value in both cases has several obvious downward steps during the optimization. That is caused by the step-by-step changes of C_{dw} to ensure better convergence. As for the constraint, a stable iteration process can be found in both cases. Through this example, the stable convergence performance of the proposed method can be well verified.

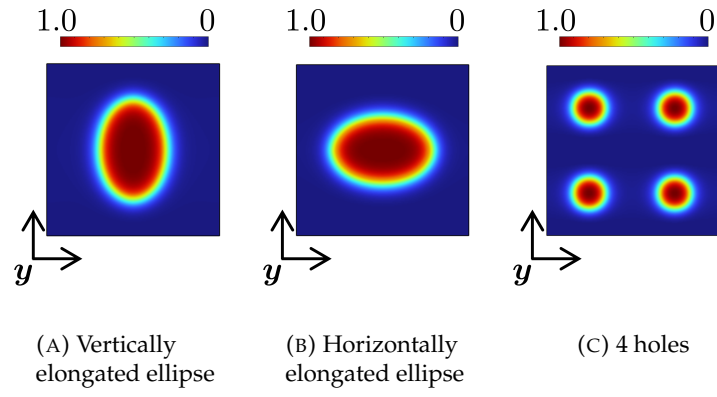


FIGURE 3.14: The initial distributions in the unit cell for the optimization

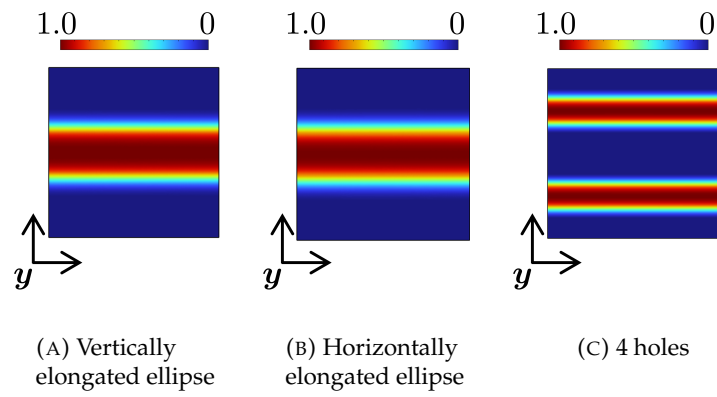


FIGURE 3.15: The obtained results for each initial distribution

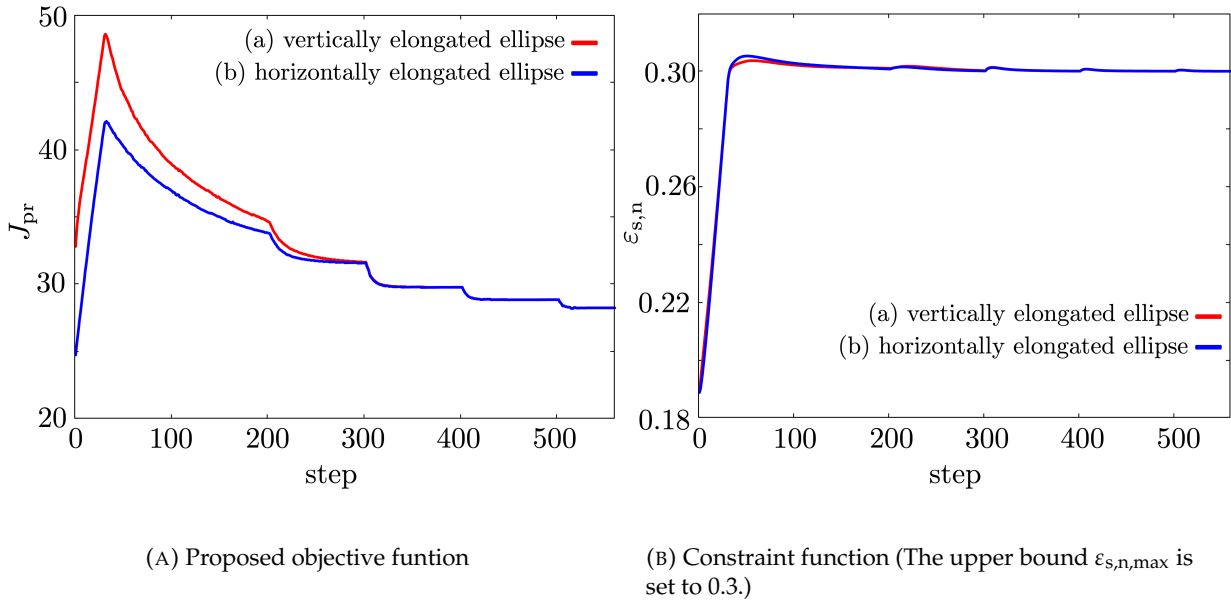


FIGURE 3.16: Plots for the objective and constraint function histories for the initial distributions

Effect on macrostructure configurations

Next, we examine the effect of different macrostructure configurations on optimized material distribution within the unit cell. In this study, we change the macroscale domain by altering the interface shape between the composite anode and the electrolyte phase. Three interface types are involved into the consideration, with their shape described by type A, type B, and type C, as shown in Eq. (3.35). Especially for type B, required parameters Q and n are set to 10^{-6} and 1, respectively. Specifically, the shape Type A refers to a vertical straight interface and Type B describes a interface represented by a sine function with the amplitude of 10^{-6} and a frequency of 1. Type C has interface that is a straight line with a gradient of $2/5$. Here, the optimization results considering the interface shape Type A has already been presented in Fig. 3.13b, and Fig. 3.17 shows the optimized microstructural configurations based on the macroscale configuration with interface shape Type B and C. From these results, it is found that when a periodic function is used as the interface, the optimal results become the same as type A, where the interface is straight in the vertical direction. From the view of the authors, this phenomenon mainly because of the periodic layout of the electrode reaction and the variations in electrode reactions in x_2 direction cancel each other out when the sensitivity is calculated. Moreover, the results obtained for type C, as shown in Eq. (3.35c), show that the distribution of the active material slightly bulges in the center of the unit cell and tends towards the bottom right. This result indicates that when the macrostructure interface is diagonal, the active material transporting electrons and the electrolyte

transporting lithium ions change their distribution to increase the diffusion coefficient in the diagonal direction. The reason why the active material does not distribute in a single diagonal line is due to the periodic conditions applied to the design variables ρ to ensure connectivity, as shown in Eq. (3.32b).

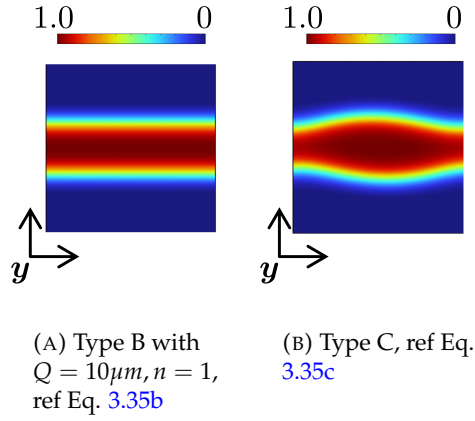


FIGURE 3.17: The obtained results for each macro scale structure

Considering two types of unit cells

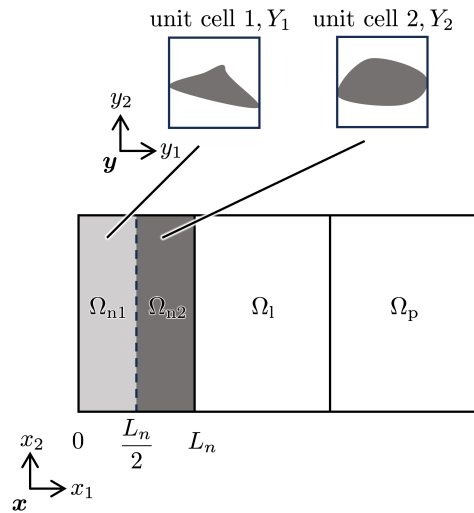


FIGURE 3.18: Schematic of two unit cells considered in the composite anode

To demonstrate the validity of the proposed method, we perform topology optimization considering two microstructures within the composite anode. As shown in Fig. 3.18, the composite anode region with a macrostructure of type A is uniformly divided into two parts, i.e., defined as Ω_{n1} and Ω_{n2} , from left to right. It is assumed that each macroscale region is respectively filled

with independently periodic layout of unit cells Y_1 and Y_2 , whose microstructural topologies are optimized via design variables ρ_1 and ρ_2 . The C-rate is set to 1.0. Furthermore, as constraints, the target average volume fraction of the active material distributed within the two unit cells is imposed to be 0.3, as follows:

$$\frac{1}{2} \left(\frac{1}{|Y_1|} \int_{Y_1} \rho_1 dY + \frac{1}{|Y_2|} \int_{Y_2} \rho_2 dY \right) - 0.3 = 0 \quad (3.37)$$

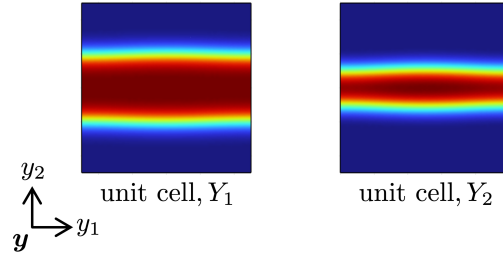


FIGURE 3.19: The obtained results for two unit cells, Y_1 and Y_2 , considered in the composite anode

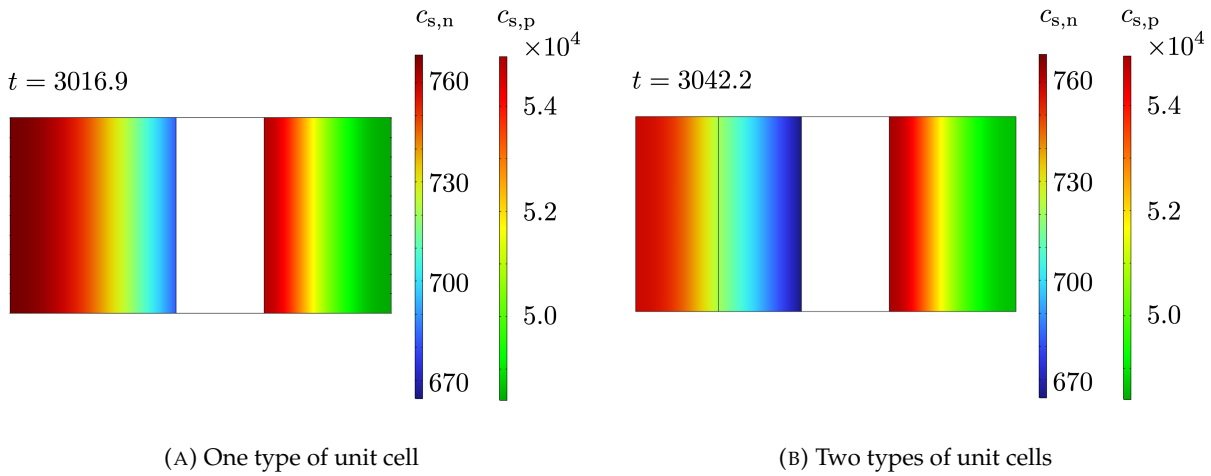


FIGURE 3.20: Concentration distributions upon termination of the calculation for the discharging process when considering one and two types of unit cells

The results are illustrated in Fig. 3.19. The volume fractions of the active material in unit cells Y_1 and Y_2 are 34.8% and 26.2%, respectively, and the active and electrolyte materials are distributed horizontally. These results indicate that regions closer to the electrode benefit from a higher volume fraction of active material. This finding aligns with the trends observed in studies on porous reactors (Alizadeh et al., 2023). The objective function value J_{pr} of the obtained result is 12.2, although the objective function value of the obtained result, assuming only one unit cell as described in Section 3.4.4, is 28.4. In other words, considering two types of unit cells yielded better outcomes.

Figure 3.20 shows the concentration distributions upon termination of the calculation for the discharging process when considering one and two types of unit cells. The concentration field within the composite anode in Fig. 3.20b is lower than in Fig. 3.20a. It reveals that considering two types of unit cells allows for a more uniform utilization of lithium within the composite anode during discharge. These trends can be attributed to the increased degrees of freedom in material representation that come with considering a greater variety of unit cells, and the proposed method is validated by the fact that the optimal structures of each unit cell can be obtained.

3.5 Conclusions

In this paper, we constructed a TO method for the design of microstructures within ASSBs, using the homogenization method. We achieved the following concluding remarks:

1. We formulated the optimization problem with a notable objective function, predicated on the hypothesis that a uniform reaction within the composite region during the discharging process leads to more effective use of the active material. Using the original function, a time integration covering the discharging and charging process is needed. However, the proposed objective function only needs an integration over the discharging process, which significantly reduces the computational cost and simplifies the adjoint equations.
2. We demonstrated the utility of the proposed objective function, which can halve the computational cost, by comparing it with the commonly used battery evaluation metrics. In the comparison, both the macrostructure and microstructure configurations were varied to compare changes to different types. The fact that the variations of both functions for configuration's changes were almost the same and the original objective function could be made smaller by minimizing the proposed objective function method, which shows the utility of the proposed objective function.
3. The sensitivity for the design of microstructures for ASSBs was derived using Lagrange and continuous adjoint methods, and the adjoint equations were denoted using strong form. Iteration histories of the objective and constraints validate the effectiveness of the derived sensitivity.
4. Optimization results show that the materials are tended to be distributed horizontally for maximizing the diffusion and conductivity efficiency horizontally. Such a tendency remains when various conditions are considered, such as different C-rates and varied initial microstructural guesses of microstructures. It is also observed that the material distribution within the unit cells changes with the variation of the macro-structural domain, tending to modify the direction of diffusion and conduction.

5. Two types of microstructures were considered in the design problem, and the optimized results within the composite anode proved the extensibility of the proposed approach. According to the obtained material layout pattern, the region closer to the electrode benefited from a higher volume fraction of active material, contributing to a better electrode reaction within the composite region.

It should be noted that the proposed method is limited to batteries driven by single ions, with no defects at the reaction interface and no dendrite growth during the charge and discharge processes. Considering as many phenomena as possible will increase the reliability of the battery design obtained by the optimization method. The authors believe that the proposed method will serve as a foundation for other researchers to establish more sophisticated battery models in the future by taking into account various realistic phenomena occurring in batteries.

Chapter 4

Data-driven topology optimization of all-solid-state batteries considering conductive additive material informed by microstructure analysis[†]

CONTENTS

4.1 Introduction	80	4.5.2 Material interpolation function profile	102
4.2 Formulation	84	4.5.3 Optimization results	110
4.2.1 Design variables settings	84	Performance evaluation of optimized material distribution: Comparison with uniform distribution	111
4.2.2 Governing equations	85	Comparison and analysis of optimal solutions obtained using different material interpolation functions	114
4.2.3 Optimization problem	89	Relation between electronic conductivity values of materials and optimized structures	116
4.3 Material interpolation scheme	92		
4.3.1 Random material distribution schemes	92	4.6 Conclusions	116
4.3.2 Estimation of normalized transport coefficient and surface area	95		
4.3.3 Algorithm for obtaining interpolation functions	98		
4.4 Numerical implementations	101		
4.5 Numerical examples	102		
4.5.1 Numerical settings	102		

[†]The work in this chapter has been published in *Structural and Multidisciplinary Optimization*, DOI: 10.1007/s00158-025-04094-9.

4.1 Introduction

All-solid-state batteries (ASSBs) have gained interest as a notable energy storage device. Compared to conventional liquid-based lithium-ion batteries, ASSBs are less flammable and have a lower risk of ignition or explosion, resulting in improved safety (Janek and Zeier, 2016). Furthermore, the use of solid electrolytes instead of liquid electrolytes increases effective energy density and enables more efficient energy storage (Wang et al., 2020; Zhang et al., 2020; Nanda et al., 2021). This technological advantage will allow for more effective use of renewable energy and increase the cruising range of electric vehicles, and as a result, this contributes to the achievement of a carbon-neutral society (Cano et al., 2018). Therefore, the development and widespread use of ASSBs is a key approach for laying the foundation for a sustainable energy system.

However, ASSBs have a drawback compared to liquid-based lithium-ion batteries, i.e., the insufficient contact area among materials composed in the composite region, resulting in a decrease of conductivity, diffusion coefficient, and electrochemical reaction rate (Ren et al., 2023; Deysher et al., 2022; Minnmann et al., 2021). To mitigate this issue, many research groups are making significant efforts in various ways, such as material development (Yong-Gun et al., 2020) and the improvement of manufacturing processes (Giraldo et al., 2021). Recently, conductive additive materials have been increasingly incorporated into the components of ASSBs to enhance electronic conductivity (Giraldo et al., 2021; Reisacher et al., 2023). As mentioned above, performance improvements and material development have been carried out, but due to the complexity of coupled reaction-transport phenomena in the electrodes, a design method for the batteries has yet to be established.

In such a battery development situation, optimization methods have the potential to improve the performance of all kinds of batteries. A typical application is the material composition optimization of battery cathode and anode materials to enhance performances including effective energy density, cycle life, and charge/discharge rate (Lv et al., 2022; Ong, 2019; Shahzad et al., 2024). Those researchers used a combined method of high-throughput experiments and numerical approaches to efficiently find optimized solutions in a short period.

Another application of the optimization method is obtaining the optimized configuration of battery devices such as the particle sizes of the active material and solid electrolyte. Many research groups have attempted to find optimal values for the properties of the materials present in the battery by combining experiments and numerical simulations to increase the contact area among materials. For example, Shi et al., 2020 performed parametric optimization for the battery performance by varying the particle sizes of the active material and solid electrolyte in the cathode based on a numerical model confirmed by experimental data. They found that an optimal particle size ratio improved the battery performance. Clausnitzer et al., 2023 developed a method to find the optimal solution based on structure-resolved simulations targeting the values of the active material fraction, the density after sintering, and the particle size as objective functions.

Moreover, they revealed the effective energy density could be improved using the functionally graded material (FGM) approach (Deng et al., 2024), wherein the volume fraction of active material is increased towards the current collector, and the volume fraction of electrolyte is increased towards the separator. That is, the increase of the contact area and appropriate material distribution by these proposed methods can increase the transport of both Li ions and electrons and charge transfer, resulting in improving power density of the battery.

However, although previously proposed optimization methods provide practical solutions, they are limited in terms of the design variables' degree of freedom and the range of feasible solutions. Thus, topology optimization (TO) is one of most promising approaches to obtain further improved and innovative design configurations.

Development of a TO method originated from structural problems (Bendsøe and Kikuchi, 1988), and it is now extended to various physics problems. Borrvall and Petersson, 2003 and Matsumori et al., 2013 have applied it to fluid and thermal fluid problems, respectively, and they obtained the optimized fluid channel layout of heat exchangers. Wadbro and Berggren, 2006 and Lu et al., 2013 have constructed a TO method for the acoustic problem and obtained an optimized acoustic horn and metasurface design, respectively.

TO has also been applied to various types of electrochemical devices, including batteries and fuel cells. Iwai et al., 2011 constructed the TO method to enhance the efficiency of solid oxide fuel cells (SOFC). Yaji et al., 2018 applied TO to improve the performance of redox flow batteries, which utilize liquid electrolytes at both the anode and cathode. The TO methods were also constructed by Lin et al., 2022 and Roy et al., 2022 to refine internal flow paths within the redox flow batteries, further increasing operational efficiency. Additionally, Mo et al., 2021 and Guo et al., 2022a proposed the TO method that can provide cooling plates design, addressing the thermal management challenges inherent to battery systems. Pejman and Najafi, 2023 presented a TO method for providing the optimized configurations of solid and compliant phases of structural battery composites (SBC) by modeling them with equivalent physical networks. SBC embodies a novel class of multifunctional materials, integrating the load-bearing capabilities of carbon fiber composites with the energy storage properties characteristic of lithium-ion batteries (Snyder et al., 2007; Snyder et al., 2008; Wong et al., 2007; Liu et al., 2009). Furthermore, Alizadeh et al., 2023 proposed a TO method for optimizing porous reactors, which concerns the spatial distribution of active and electrolyte material volume fractions.

As for ASSBs, Ishida et al., 2024 proposed a TO method based on a homogenization approach. Their study derives sensitivities using the Lagrange adjoint method for the discharge process, which is inherently time-dependent, and demonstrates that the proposed optimization framework can identify the optimal microstructural configuration of active and electrolyte materials within a unit cell of the composite anode. However, fabricating such microstructural configurations is extremely challenging with current manufacturing technologies. Thus, a more macroscopic optimization approach is needed to ensure practical feasibility.

Therefore, this paper is primarily devoted to the development of a TO method that systematically determines the optimized macroscopic material distribution configuration for ASSBs. Specifically, the proposed method optimizes the volume fraction distributions of electrolytes, active materials, and conductive additives in the composite anode so as to maximize the functional contributions of each material during the discharge process.

One of the key issues in macroscopic-level optimization is the material interpolation function. While conventional topology optimization typically employs the solid isotropic material with penalization (SIMP) method to penalize intermediate densities (Bendsøe, 1989; Bendsøe and Sigmund, 1999), this approach is not suitable for battery optimization where intermediate material compositions play a crucial role. Instead, we require material interpolation functions that accurately reflect the physicochemical properties of mixed material states, as these intermediate compositions are fundamental to battery performance.

The Bruggeman model has been widely used in battery analysis to predict effective transport coefficients based on material volume fractions (Chung et al., 2013). This model interpolates the material's macroscopic properties with a function corresponding to the penalty parameter p of 1.5 in the SIMP method, assuming that the material particles are spherical (Tjaden et al., 2016). The unit cell size used in the Bruggeman model, in which transport is evaluated, is assumed to be sufficiently large relative to the particle size, i.e., approximately the size of the composite anode region. However, in this paper, the material distributions within the composite anode vary on a scale smaller than the size of the composite anode region, and this fact indicates the use of the Bruggeman model is not appropriate in the proposed optimization method. In other words, the size of the unit cell should be significantly smaller than the composite anode region.

Thus, this paper develops the material interpolation function with microstructure analysis, i.e., that randomly distributes materials in a pseudo unit cell, evaluates their properties, and uses the least-squares method to obtain an approximated function from the obtained datasets. Next, the optimization problem is formulated to minimize the ratio of overvoltage to current during the discharge process, which serves as an indicator of resistance in the battery system. The sensitivity is based on the sensitivity derivation proposed by Ishida et al., 2024. After that, the optimization algorithm is developed. Finally, numerical examples demonstrate the effectiveness of the proposed method.

The rest of paper is organized as follows. Section 4.2 describes the mathematical formulation of governing equations for ASSBs and the optimization problem for minimizing the overvoltage. Section 4.3 presents the approximation method for the material interpolation functions. In Section 4.4, the optimization algorithm is detailed. After that, Section 4.5 provides several numerical examples to confirm the validity of the proposed method. Finally, the conclusions of this paper are presented in Section 4.6.

The variables used in this paper are summarized in Table 4.1.

TABLE 4.1: List of variables used in the paper

Variable	Description	Unit
$\varepsilon_{a,n}$	Volume fraction of active material in the composite anode	–
ε_c	Volume fraction of conductive additive in the composite anode	–
$\varepsilon_{e,n}$	Volume fraction of electrolyte material in the composite anode	–
c_n	Li concentration in the composite anode	mol/m ³
c_p	Li concentration in the composite cathode	mol/m ³
ϕ_n	Electrical potential of active material in the composite anode	V
ϕ_p	Electrical potential of active material in the composite cathode	V
ϕ_e	Electrical potential of electrolyte material	V
D_n^{eff}	Li diffusion coefficient of active material in the composite anode	m ² /s
D_p^{eff}	Li diffusion coefficient of active material in the composite cathode	m ² /s
σ_n^{eff}	Electronic conductivity in the composite anode	S/m
σ_p^{eff}	Electronic conductivity in the composite cathode	S/m
κ^{eff}	Li-ion conductivity	S/m
j^{Li}	Charge-transfer current density	A/m ³
η	Overvoltage	V
a_s^{eff}	Active surface area per electrode unit volume for charge-transfer reaction	m ² /m ³
I	Current	A
Q_{cell}	Capacity	C
J	Objective function	V
$a_1 - a_2$	Adjoint variables	V · m/mol
$a_3 - a_5$	Adjoint variables	m/(S · s)
J'_1	Sensitivity for $\varepsilon_{a,n}$	V
J'_2	Sensitivity for ε_c	V
ρ	Randomly generated material distribution in the unit cell [0 : 1]	–
$C_1 - C_3$	Dimensionless scalar fields defined in the unit cell	–
$q_1 - q_3$	Dimensionless fluxes of C_1 , C_2 , and C_3	–
K	Dimensionless transport coefficient	–
$Q_1 - Q_3$	Dimensionless fluxes of C_1 , C_2 , and C_3 on Γ_1 , Γ_2 , and Γ_3	–
$W_1 - W_3$	Approximated dimensionless transport coefficients	–

4.2 Formulation

This section describes the mathematical formulation for the optimization used in this study. First, Section 4.2.1 describes the battery configuration and design variables to be optimized. Section 4.2.2 then presents the analytical model of the ASSBs and clarifies the governing equations to be solved. Finally, Section 4.2.3 formulates an optimization problem aimed at minimizing the internal losses of the battery.

4.2.1 Design variables settings

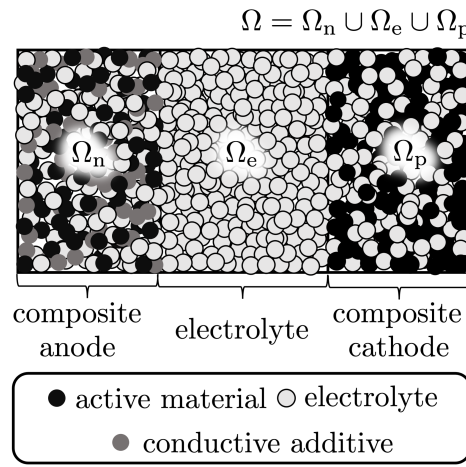


FIGURE 4.1: Model of all-solid-state-batteries

Figure 4.1 shows the battery configuration that is the analysis domain of this research. The battery consists of three regions: composite anode (negative electrode) Ω_n , electrolyte Ω_e , and composite cathode (positive electrode) Ω_p . The composite cathode is a composite region of active and electrolyte materials, while the composite anode is the composite region of active, electrolyte, and conductive additive materials. In the electrolyte, only the electrolyte material exists. The three materials composing the battery have different roles. The active material stores Li that moves between the cathode and anode during charging and discharging, and the electrolyte carries Li ions. The conductive additive material is a highly conductive material added to increase the electronic conductivity of the composite anode.

The design goal of this study is to find the optimal distributions of the three materials in the composite anode Ω_n . That is, the volume fraction of each material is assumed to be not constant in the composite anode, but depends on the position coordinates, and then the volume fractions of these materials are set to the design variables. Let $\varepsilon_{a,n}$, $\varepsilon_{e,n}$, and ε_c be the volume fractions of the active, electrolyte, and conductive additive materials in the composite anode, respectively. Here, this paper assumes that there are no voids in the composite anode.

4.2.2 Governing equations

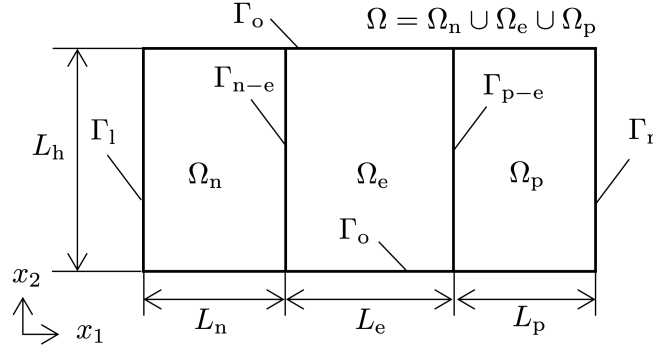


FIGURE 4.2: Domains and boundaries definitions

The analysis in this study is conducted in a two-dimensional domain, as shown in Fig. 4.2. As mentioned in the previous section, the computational domain Ω consists of three regions: composite anode Ω_n , composite cathode Ω_p , and electrolyte Ω_e , with thicknesses L_n , L_p , and L_e , respectively. The height is L_h . The left boundary of the composite anode is represented as Γ_l , and the right boundary of the composite cathode as Γ_r . The interfaces between the composite anode and the electrolyte and between the composite cathode and the electrolyte are denoted by Γ_{n-e} and Γ_{p-e} , respectively.

Next, time and spatial scales are defined within the computational domain, as shown in the following equations:

$$t \in \mathcal{T}[0, t_{\max}] \subseteq \mathbb{R}_{\geq 0}, \quad (4.1)$$

$$\mathbf{x} = (x_1, x_2) \in \Omega \subseteq \mathbb{R}^2. \quad (4.2)$$

where $t_{\max}$ is the terminated time of the discharge analysis. The termination condition for the analysis of the discharge process to determine $t_{\max}$ is described in detail in the latter part of this section.

There are five variables required in this analysis model. Four of those are the Li concentrations, c_n and c_p , and electrical potentials, ϕ_n and ϕ_p , defined in each composite region. The fifth variable is the electrical potential ϕ_e in the electrolyte phase, defined in all regions where the electrolyte is present. In this study, as in Ishida et al., 2024, the transport number of Li ions is assumed to be unity (KANNO, 2023), and Li and electron diffusion follows Fick's law (Fuller and Harb, 2018). The governing equations, initial conditions and boundary conditions to be solved are described

as follows:

$$\varepsilon_{a,n} \frac{\partial c_n}{\partial t} = \nabla \cdot \left(D_n^{\text{eff}} \nabla c_n \right) - \frac{j^{\text{Li}}}{F} \quad \text{in } \mathcal{T} \times \Omega_n, \quad (4.3)$$

$$\varepsilon_{a,p} \frac{\partial c_p}{\partial t} = \nabla \cdot \left(D_p^{\text{eff}} \nabla c_p \right) - \frac{j^{\text{Li}}}{F} \quad \text{in } \mathcal{T} \times \Omega_p, \quad (4.4)$$

$$\nabla \cdot \left(\sigma_n^{\text{eff}} \nabla \phi_n \right) = j^{\text{Li}} \quad \text{in } \mathcal{T} \times \Omega_n, \quad (4.5)$$

$$\nabla \cdot \left(\sigma_p^{\text{eff}} \nabla \phi_p \right) = j^{\text{Li}} \quad \text{in } \mathcal{T} \times \Omega_p, \quad (4.6)$$

$$\nabla \cdot \left(\kappa^{\text{eff}} \nabla \phi_e \right) = -j^{\text{Li}} \quad \text{in } \mathcal{T} \times \Omega, \quad (4.7)$$

$$c_n = c_{n,\text{max}} \times \text{SOC}_{n,\text{max}} \quad \text{in } \mathcal{T}(0) \times \Omega_n, \quad (4.8)$$

$$c_p = c_{p,\text{max}} \times \text{SOC}_{p,\text{min}} \quad \text{in } \mathcal{T}(0) \times \Omega_p, \quad (4.9)$$

$$D_n^{\text{eff}} \nabla c_n \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{l,o,n-e}, \quad (4.10)$$

$$D_p^{\text{eff}} \nabla c_p \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{r,o,p-e}, \quad (4.11)$$

$$\phi_n = 0 \quad \text{on } \mathcal{T} \times \Gamma_l, \quad (4.12)$$

$$\sigma_n^{\text{eff}} \nabla \phi_n \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{o,n-e}, \quad (4.13)$$

$$\sigma_p^{\text{eff}} \nabla \phi_p \cdot \mathbf{n} = -\frac{I}{A_g} \quad \text{on } \mathcal{T} \times \Gamma_r, \quad (4.14)$$

$$\sigma_p^{\text{eff}} \nabla \phi_p \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{o,p-e}, \quad (4.15)$$

$$\kappa^{\text{eff}} \nabla \phi_e \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{l,r,\text{out}}, \quad (4.16)$$

where $\varepsilon_{a,p}$ is the volume fraction of active material in the composite cathode and $\mathbf{n}$ is the outward normal vector. $c_{n,\text{max}}$ and $c_{p,\text{max}}$ are the upper limits of the Li concentration that can exist in the composite anode and cathode material, respectively. $\text{SOC}_{n,\text{max}}$ and $\text{SOC}_{p,\text{min}}$ are the upper and lower limits of the state of charge in the composite anode and cathode, respectively. A_g is the area of the current collector. j^{Li} and η are the reaction current density and overvoltage, respectively, and are defined as follows:

$$j^{\text{Li}} = a_s^{\text{eff}} i_0 \left[\exp \left(\frac{0.5F}{RT} \eta \right) - \exp \left(-\frac{0.5F}{RT} \eta \right) \right], \quad (4.17)$$

$$\eta = \begin{cases} \phi_n - \phi_e - U_n(c_n) & \text{in } \Omega_n \\ 0 & \text{in } \Omega_e \\ \phi_p - \phi_e - U_p(c_p) & \text{in } \Omega_p, \end{cases} \quad (4.18)$$

where i_0 is the exchange current density, and a_s^{eff} represents the effective surface area per unit volume. Then, F , R , and T are the Faraday constant, gas constant, and temperature, respectively. In this study, the temperature is assumed to be constant. U_n and U_p are functions that determine the standard equilibrium electrode potential, which depends on the type of material used in each composite region and the Li concentration.

As shown in Eq. 4.14, the condition given to the system is assumed to be a constant current. The magnitude of the given current is related to the capacity of the battery, as follows:

$$I = \frac{Q_{\text{cell}}}{3600[\text{s}]} C_{\text{rate}}, \quad (4.19)$$

where C_{rate} is a dimensionless parameter that determines the magnitude of the battery discharge. By the definition, C_{rate} of 1 represents the magnitude of the current at which the battery is fully discharged in one hour. This study defines the capacity of the battery Q_{cell} based on the amount of active material in the composite anode, as shown in the following equation:

$$Q_{\text{cell}} = c_{n,\text{max}}(\text{SOC}_{n,\text{max}} - \text{SOC}_{n,\text{min}}) \tilde{\epsilon}_{a,n} V_n F, \quad (4.20)$$

where V_n is the volume of the composite anode region. $\text{SOC}_{n,\text{min}}$ is the lower limit of the state of charge in the composite anode, and $\tilde{\epsilon}_{a,n}$ is the average volume fraction of active material in the composite anode. Then, the initial conditions for the Li concentrations of the composite anode and cathode are given as Eqs. (4.8) and (4.9), respectively. Namely, the initial conditions for this study are set to be fully charged in order to calculate the discharge process. It should be noted that the proposed method allows for an arbitrary initial state of the battery.

Next, we explain the termination conditions for the discharge analysis. As mentioned above, the magnitude of the current is given so that the battery should discharge in certain time. However, due to the non-linearity of the governing equations, the time taken for the discharge is not an explicit value, and can change during the optimization process. Therefore, in this study, the discharge calculation is terminated when the terminal voltage, which decreases during the discharge process, falls below a specific threshold value $V_{\text{terminate}}$, as shown in the following equation:

$$\frac{\int_{\Gamma_r} \phi_p \, d\Gamma}{\int_{\Gamma_r} d\Gamma} \leq V_{\text{terminate}}. \quad (4.21)$$

Finally, we explain the material interpolation method. That is, the following formulations reveal which variables depend on which material and how they depend on the volume fraction of the material. In the analytical model in this paper, there are two types of effective coefficients that require material interpolation, i.e., the transport coefficient and the surface area of active material per unit volume in the composite anode, where the design variables are defined, and the composite cathode. For the transport coefficient, there are five effective coefficients in the governing equations. The diffusion coefficients of Li in the composite anode and composite cathode are functions of the volume fractions of active material, $\epsilon_{a,n}$ and $\epsilon_{a,p}$, in each domain. The electronic conductivity in the composite anode depends on the volume fractions of both active material and conductive additive material, $\epsilon_{a,n}$ and ϵ_c . The electronic conductivity in the composite cathode depends on the volume fraction of the active material, $\epsilon_{a,p}$. The Li-ion conductivity in the electrolyte depends on the volume fraction of the electrolyte in each region. Then, the surface area of active

material per unit volume also depends on the volume fraction of active material within each domain and the particle size of the active material. For simplicity, this study assumes the particle size is constant in the composite region. These relationships between the volume fractions and effective material properties can be expressed in the following equations:

$$D_n^{\text{eff}} = D_n H_1(\varepsilon_{a,n}(\mathbf{x})), \quad (4.22)$$

$$D_p^{\text{eff}} = D_p H_1(\varepsilon_{a,p}), \quad (4.23)$$

$$\sigma_n^{\text{eff}} = \sigma_n H_2(\varepsilon_{a,n}(\mathbf{x}), \varepsilon_c(\mathbf{x})), \quad (4.24)$$

$$\sigma_p^{\text{eff}} = \sigma_p H_1(\varepsilon_{a,p}), \quad (4.25)$$

$$\kappa^{\text{eff}} = \begin{cases} \kappa H_1(\varepsilon_{e,n}(\mathbf{x})) & \text{in } \Omega_n \\ \kappa H_1(\varepsilon_{e,e}) & \text{in } \Omega_e \\ \kappa H_1(\varepsilon_{e,p}) & \text{in } \Omega_p, \end{cases} \quad (4.26)$$

$$a_s^{\text{eff}} = \begin{cases} \frac{A(\varepsilon_{a,n}(\mathbf{x}))}{L} & \text{in } \Omega_n \\ \frac{A(\varepsilon_{a,p})}{L} & \text{in } \Omega_p, \end{cases} \quad (4.27)$$

where $\varepsilon_{e,e}$ and $\varepsilon_{e,p}$ represent the volume fractions of electrolyte materials in electrolyte phase and the composite cathode, respectively. H_1 , H_2 and A are the dimensionless interpolation functions for the transport coefficients and surface area proposed in this paper, respectively. The details of these functions are described in Section 4.3. L is the length of the unit cell defined in Section 4.3.1. D_n and D_p are the Li diffusion coefficient of active material in the composite anode and cathode, respectively. σ_n and σ_p are the conductivity of active material in the composite anode and cathode, respectively. Moreover, κ is the ionic conductivity of electrolyte material. To clarify our paper description, it should be remarked that $\varepsilon_{a,n}$, ε_c , and $\varepsilon_{e,n}$ depend on the coordinates of the position, while $\varepsilon_{a,p}$, $\varepsilon_{e,p}$, and $\varepsilon_{e,e}$ are constants. This is because the design variable is set to the volume fraction distribution only in the composite anodes in this paper.

4.2.3 Optimization problem

In this section, we formulate an optimization problem aiming at minimizing the energy loss during the discharge process. For simplicity of notation, the following variables are defined:

$$\tilde{c}_p = \frac{\int_{\Omega_p} c_p d\Omega}{\int_{\Omega_p} c_{p,\max} d\Omega}, \quad (4.28)$$

$$\tilde{\phi}_s = \frac{\int_{\Gamma_r} \phi_s d\Gamma}{\int_{\Gamma_r} d\Gamma}, \quad (4.29)$$

$$\tilde{\varepsilon}_{a,n} = \frac{\int_{\Omega_n} \varepsilon_{a,n} d\Omega}{\int_{\Omega_n} d\Omega}, \quad (4.30)$$

$$\tilde{\varepsilon}_c = \frac{\int_{\Omega_n} \varepsilon_c d\Omega}{\int_{\Omega_n} d\Omega}, \quad (4.31)$$

where $\tilde{c}_p$ is the average Li concentration in the composite anode, and $\tilde{\phi}_s$ is the average potential of the active material at the rightmost boundary. Moreover, $\tilde{\varepsilon}_{a,n}$ and $\tilde{\varepsilon}_c$ are the average volume fractions of active material and conductive additive material in the composite anode, respectively. Using these variables, the optimization problem is formulated as follows:

$$\inf_{\varepsilon_{a,n}, \varepsilon_c} J = \frac{\int_{\mathcal{T}} (U_p(\tilde{c}_p) - \tilde{\phi}_s) dt}{t_{\max} I}, \quad (4.32)$$

$$s.t. \quad \underline{\varepsilon}_{a,n} \leq \varepsilon_{a,n} \leq \overline{\varepsilon}_{a,n}, \quad (4.33)$$

$$\underline{\varepsilon}_c \leq \varepsilon_c \leq \overline{\varepsilon}_c, \quad (4.34)$$

$$\varepsilon_{a,n}(\mathbf{x}) + \varepsilon_c(\mathbf{x}) + \varepsilon_{e,n}(\mathbf{x}) = 1, \quad (4.35)$$

$$\frac{\tilde{\varepsilon}_{a,n}}{\overline{V}_a} - 1 \leq 0, \quad (4.36)$$

$$\frac{\tilde{\varepsilon}_c}{\overline{V}_c} - 1 \leq 0. \quad (4.37)$$

The objective function is set as the ratio of the overvoltage during the discharge process and the current. The overvoltage is the difference of the standard electrode potential based on the average Li concentration in the cathode and the terminal voltage. Namely, the overvoltage corresponds to the amount of energy required to drive the battery reaction, and a small absolute value of this value means that the energy loss inside the battery is small (Alizadeh et al., 2023). On the other hand, a higher current draw is preferable, given the performance requirements of the battery. That is, minimizing the objective function aims to minimize the overvoltage and maximize the current at the same time.

Since this study considers only the discharge process, the overvoltage in the composite cathode is negative. Thus, it is converted to a positive value by multiplying by -1. It should be noted that the sign of the objective function changes when the charging process is considered. The design

variables are $\varepsilon_{a,n}$ and ε_c . As mentioned in Section 4.2.1, the objective of this study is to obtain the distribution of the volume fractions of active material, electrolyte, and conductive additive in the composite anode. Since the presence of voids does not positively affect the optimized solution, we assume that there are no voids and that Eq. (4.35) is satisfied. Therefore, the number of the design variables can be reduced. Here, $\overline{\varepsilon_{a,n}}$ and $\overline{\varepsilon_c}$ are the upper bounds of the volume fraction of active material and conductive additive, respectively, and $\underline{\varepsilon_{a,n}}$ and $\underline{\varepsilon_c}$ are the lower limits of those, respectively. Moreover, as shown in Eq. (4.36) and Eq. (4.37), the volume constraints for each material with the upper limits $\overline{V}_a$ and $\overline{V}_c$ are taken into account.

Next, we derive the sensitivity of the objective function formulated above using the Lagrange and continuous adjoint methods. First, the Lagrangian is defined as follows:

$$\begin{aligned} \mathcal{L} = & J + \int_{\mathcal{T}} \int_{\Omega_n} a_1 \left(\varepsilon_{a,n} \frac{\partial (c_n)}{\partial t} - \nabla \cdot (D_n^{\text{eff}} \nabla c_n) + \frac{j^{\text{Li}}}{F} \right) d\Omega dt \\ & + \int_{\mathcal{T}} \int_{\Omega_p} a_2 \left(\varepsilon_{a,p} \frac{\partial (c_p)}{\partial t} - \nabla \cdot (D_p^{\text{eff}} \nabla c_p) + \frac{j^{\text{Li}}}{F} \right) d\Omega dt \\ & + \int_{\mathcal{T}} \int_{\Omega_n} a_3 \left(\nabla \cdot (\sigma_n^{\text{eff}} \nabla \phi_n) - j^{\text{Li}} \right) d\Omega dt \\ & + \int_{\mathcal{T}} \int_{\Omega_p} a_4 \left(\nabla \cdot (\sigma_p^{\text{eff}} \nabla \phi_p) - j^{\text{Li}} \right) d\Omega dt \\ & + \int_{\mathcal{T}} \int_{\Omega} a_5 \left(\nabla \cdot (\kappa_m^{\text{eff}} \nabla \phi_{e,m}) + j^{\text{Li}} \right) d\Omega dt, \end{aligned} \quad (4.38)$$

where a_1, a_2, a_3, a_4 , and a_5 are the adjoint variables. This Lagrangian can be differentiated by the design variables and transformed using the chain rule as follows:

$$\begin{aligned} \frac{d\mathcal{L}}{d\varepsilon_{[\cdot]}} = & \frac{\partial \mathcal{L}}{\partial \varepsilon_{[\cdot]}} + \frac{\partial \mathcal{L}}{\partial c_n} \frac{\partial c_n}{\partial \varepsilon_{[\cdot]}} + \frac{\partial \mathcal{L}}{\partial c_p} \frac{\partial c_p}{\partial \varepsilon_{[\cdot]}} \\ & + \frac{\partial \mathcal{L}}{\partial \phi_n} \frac{\partial \phi_n}{\partial \varepsilon_{[\cdot]}} + \frac{\partial \mathcal{L}}{\partial \phi_p} \frac{\partial \phi_p}{\partial \varepsilon_{[\cdot]}} + \frac{\partial \mathcal{L}}{\partial \phi_e} \frac{\partial \phi_e}{\partial \varepsilon_{[\cdot]}}, \end{aligned} \quad (4.39)$$

where $\varepsilon_{[\cdot]}$ represents $\varepsilon_{a,n}$ and ε_c . Since it is difficult to derive $\frac{\partial c_n}{\partial \varepsilon_{[\cdot]}}$, $\frac{\partial c_p}{\partial \varepsilon_{[\cdot]}}$, $\frac{\partial \phi_n}{\partial \varepsilon_{[\cdot]}}$, $\frac{\partial \phi_p}{\partial \varepsilon_{[\cdot]}}$, and $\frac{\partial \phi_e}{\partial \varepsilon_{[\cdot]}}$, we aim to find the distribution of adjoint variables that satisfy the conditions $\frac{\partial \mathcal{L}}{\partial c_n} = 0$, $\frac{\partial \mathcal{L}}{\partial c_p} = 0$, $\frac{\partial \mathcal{L}}{\partial \phi_n} = 0$,

$\frac{\partial \mathcal{L}}{\partial \phi_p} = 0$, and $\frac{\partial \mathcal{L}}{\partial \phi_e} = 0$ as the following equations show:

$$-\varepsilon_{a,n} \frac{\partial a_1}{\partial t} = \nabla \cdot (D_n^{\text{eff}} \nabla a_1) + \tilde{\eta} M \frac{\partial U_n}{\partial c_n} \quad \text{in } \mathcal{T} \times \Omega_n, \quad (4.40)$$

$$-\varepsilon_{a,p} \frac{\partial a_2}{\partial t} = \nabla \cdot (D_p^{\text{eff}} \nabla a_2) + \tilde{\eta} M \frac{\partial U_p}{\partial c_p} - \frac{\partial U_p}{\partial \tilde{c}_p} \frac{1}{L_h c_{p,\max} t_{\max} I} \quad \text{in } \mathcal{T} \times \Omega_p, \quad (4.41)$$

$$\nabla \cdot (\sigma_n^{\text{eff}} \nabla a_3) = -\tilde{\eta} M \quad \text{in } \mathcal{T} \times \Omega_n, \quad (4.42)$$

$$\nabla \cdot (\sigma_p^{\text{eff}} \nabla a_4) = -\tilde{\eta} M \quad \text{in } \mathcal{T} \times \Omega_p, \quad (4.43)$$

$$\nabla \cdot (\kappa^{\text{eff}} \nabla a_5) = \tilde{\eta} M \quad \text{in } \mathcal{T} \times \Omega, \quad (4.44)$$

$$a_1 = 0 \quad \text{in } \mathcal{T}(t_{\max}) \times \Omega_n, \quad (4.45)$$

$$a_2 = 0 \quad \text{in } \mathcal{T}(t_{\max}) \times \Omega_p, \quad (4.46)$$

$$D_n^{\text{eff}} \nabla a_1 \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{l,o,n-e}, \quad (4.47)$$

$$D_p^{\text{eff}} \nabla a_2 \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{r,o,p-e}, \quad (4.48)$$

$$a_3 = 0 \quad \text{on } \mathcal{T} \times \Gamma_l, \quad (4.49)$$

$$\sigma_n^{\text{eff}} \nabla a_3 \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{o,n-e}, \quad (4.50)$$

$$\sigma_p^{\text{eff}} \nabla a_4 \cdot \mathbf{n} = -\frac{1}{L_h t_{\max} I} \quad \text{on } \mathcal{T} \times \Gamma_r, \quad (4.51)$$

$$\sigma_p^{\text{eff}} \nabla a_4 \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{o,p-e}, \quad (4.52)$$

$$\kappa^{\text{eff}} \nabla a_5 \cdot \mathbf{n} = 0 \quad \text{on } \mathcal{T} \times \Gamma_{l,r,o}, \quad (4.53)$$

$$M = \frac{\partial j^{\text{Li}}}{\partial \eta}, \quad (4.54)$$

$$\tilde{\eta} = \begin{cases} \frac{a_1}{F} - a_3 + a_5 & \text{in } \Omega_n \\ 0 & \text{in } \Omega_e \\ \frac{a_2}{F} - a_4 + a_5 & \text{in } \Omega_p. \end{cases} \quad (4.55)$$

Finally, the sensitivities can be derived as:

$$\begin{aligned} \frac{d\mathcal{L}}{d\varepsilon_{a,n}} &= \frac{\partial \mathcal{L}}{\partial \varepsilon_{a,n}} = \int_{\Omega_n} \left(\frac{\partial J}{\partial \varepsilon_{a,n}} - a_1 c_n|_{t=0} \right) d\Omega + \int_{\mathcal{T}} \int_{\Omega_n} \left(-c_n \frac{\partial a_1}{\partial t} \right. \\ &\quad + \frac{\partial D_n^{\text{eff}}}{\partial \varepsilon_{a,n}} \nabla a_1 \cdot \nabla c_n - \frac{\partial \sigma_n^{\text{eff}}}{\partial \varepsilon_{a,n}} \nabla a_3 \cdot \nabla \phi_n \\ &\quad \left. - \frac{\partial \kappa^{\text{eff}}}{\partial \varepsilon_{a,n}} \nabla a_5 \cdot \nabla \phi_e + \left(\frac{a_1}{F} - a_3 + a_5 \right) \frac{\partial j^{\text{Li}}}{\partial \varepsilon_{a,n}} \right) d\Omega dt \\ &:= \int_{\Omega_n} J'_1 d\Omega, \end{aligned} \quad (4.56)$$

$$\begin{aligned}
\frac{d\mathcal{L}}{d\varepsilon_c} &= \frac{\partial\mathcal{L}}{\partial\varepsilon_c} = \int_{\mathcal{T}} \int_{\Omega_n} \left(-\frac{\partial\sigma_n^{\text{eff}}}{\partial\varepsilon_c} \nabla a_3 \cdot \nabla \phi_n - \frac{\partial\kappa^{\text{eff}}}{\partial\varepsilon_c} \nabla a_5 \cdot \nabla \phi_e \right) d\Omega dt \\
&:= \int_{\Omega_n} J'_2 d\Omega,
\end{aligned} \tag{4.57}$$

The details of the sensitivity derivation are noted in the Appendix of Ishida et al., 2024.

4.3 Material interpolation scheme

This section explains the derivation scheme of functions H_1 and H_2 , related to the transport coefficients that depend on the volume fraction, and function A , which is associated with surface area, both derived from microstructure analysis. Since the problem addressed in this study deals with the distribution of the material volume fraction, it is necessary to utilize interpolation functions that accurately reflect actual physical phenomena when interpolating effective properties at intermediate densities. The TO method for ASSBs proposed by Ishida et al., 2024 was based on the homogenization method, which assumes that the unit cells are arranged periodically to calculate macroscopic properties. However, as Kimura, 2023 illustrated, the actual configuration of ASSBs has a nonuniform and not periodical material distribution, making it difficult to utilize the homogenization method.

In this study, spherical materials are randomly distributed in a predefined three-dimensional domain to represent the nonuniform material distribution, and the transport coefficient value in three directions and the surface area for the material distribution are evaluated. This procedure is repeated a certain number of times to obtain datasets, and the interpolation functions are obtained by applying the least-squares method to these datasets. Moreover, the presence of Ersatz material is considered to ensure that the derived interpolation functions do not adversely affect the optimization process. It should be noted that this procedure is conducted before the optimization process performs as Section 4.4 denotes.

In Section 4.3.1, the definition of the computational domain and the representation of the material in the microstructure are explained. Next, Section 4.3.2 describes how the transport coefficients in the three directions and the magnitude of the surface area are evaluated. Then, Section 4.3.3 describes the algorithm for obtaining the interpolation function and the consideration of Ersatz material (Allaire et al., 1997).

4.3.1 Random material distribution schemes

Figure 4.3 shows the computational domain for the microstructure analysis. In this study, the variables are defined as dimensionless ones to generalize this method. In other words, the size of the cell is set to a dimensionless unit cell with the size of 1 based on the original cell size L [m].

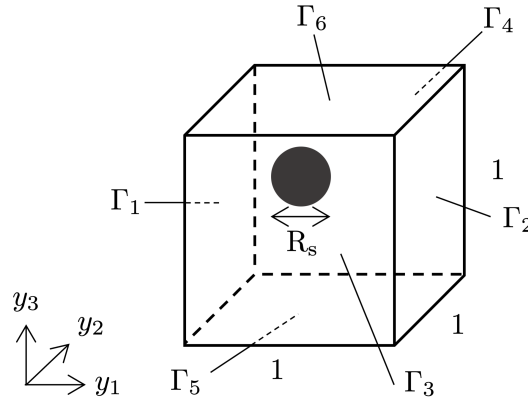


FIGURE 4.3: Model of the microstructure analysis

The spatial coordinates are defined by y_1, y_2 , and y_3 , as follows:

$$\mathbf{y} = (y_1, y_2, y_3) \in \mathbf{Y} \subseteq \mathbb{R}^3. \quad (4.58)$$

To accurately evaluate the transportation coefficients and surface area of battery materials with inherently three-dimensional characteristics, such as spherical particles, the analysis domain used for constructing interpolation functions is defined in three-dimensional space. When finding $H_1(\varepsilon)$ and $A(\varepsilon)$, which depend on the volume fraction of a single type material, a material distribution with a log-normally distributed diameter is randomly generated in this space, and when finding $H_2(\varepsilon_a, \varepsilon_c)$, which depends on the volume fraction of both active and conductive additive materials, materials with two different diameters and material properties are randomly arranged.

First, the random material generation scheme for single material type is explained. To represent the presence or absence of material, a normalized density function $\rho(\mathbf{y}) \in [0, 1]$ is defined, which takes the value 1 in regions where the material is present and 0 in void regions. The following describes how to obtain a random ρ distribution.

Based on an arbitrary random function, we obtain a set of sphere center coordinates $[\tilde{y}_{1,1}, \tilde{y}_{2,1}, \tilde{y}_{3,1}, \dots, \tilde{y}_{1,g}, \tilde{y}_{2,g}, \tilde{y}_{3,g}]$ with values between 0 and 1. Here, g is the number of spheres to be generated. Moreover, $R_{s,m} (m = 1, 2, \dots, g)$ is sampled from a normal distribution on a logarithmic scale, with a mean of $\log_{10} R_s$ and a standard deviation of s , in accordance with experimental observations (Ebner et al., 2014). Here, R_s is the mean ratio of the actual material diameters of the electrolyte and active materials to the size of the unit cell L , and the number of spheres g is determined by $\hat{\varepsilon}_t / (1/6\pi R_s^3)$. The parameter $\hat{\varepsilon}_t$ represents the approximate target volume set to 1.2 times the actual target volume $\hat{\varepsilon}$ to account for overlapping regions. Based on this dataset, a

density function $\bar{\rho}$ that takes a value of 1 inside the sphere is defined as follows:

$$\bar{\rho} = \begin{cases} 1 & \left(\text{if } (y_1 - \tilde{y}_{1,m})^2 + (y_2 - \tilde{y}_{2,m})^2 + (y_3 - \tilde{y}_{3,m})^2 \leq \left(\frac{R_{s,m}}{2} \right)^2 \right) \\ 0 & \text{(others)}, \end{cases} \quad (4.59)$$

where m is $(1, 2, \dots, g)$. Here, $\bar{\rho}$ does not have a smooth isosurface in the discretized space for numerical analysis. Therefore, $\bar{\rho}$ is smoothed using a Helmholtz filter, as shown in the following equations:

$$-R_s^2 \nabla_Y^2 \hat{\rho} + \hat{\rho} = \bar{\rho} \quad \text{in } Y, \quad (4.60)$$

$$\nabla_Y \hat{\rho} \cdot \mathbf{n} = 0 \quad \text{on } \Gamma_{1,2,3,4,5,6}, \quad (4.61)$$

where ∇_Y is the gradient in space Y and R_s is the parameter related to the filter radius. Next, the smoothed $\hat{\rho}$ is binarized using the following smoothed Heaviside function to represent the presence or absence of material:

$$\rho = \frac{\tanh\{\beta_Y(\hat{\rho} - 0.5)\} + \tanh(0.5\beta_Y)}{2 \tanh(0.5\beta_Y)}, \quad (4.62)$$

where β_Y is the steepness of the smoothed Heaviside function. Figure 4.4 illustrates the several material distributions generated using the abovementioned method. The implemented code for this material generation method is presented in Appendix B.1.

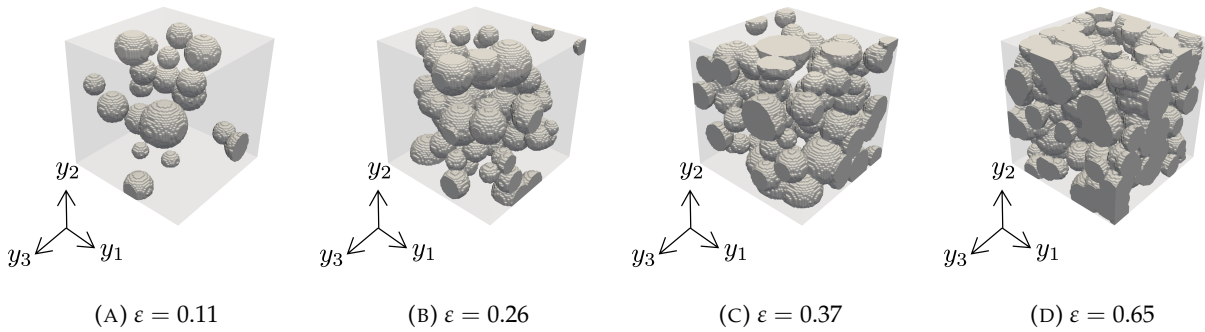


FIGURE 4.4: Randomly generated material distributions for single type of material in a unit cell with R_s of 0.2 and a standard deviation s of 0.10

Next, we explain the random material generation scheme for two types of materials within the unit cell for deriving the function $H_2(\varepsilon_a, \varepsilon_c)$. To represent the presence or absence of active and conductive additive materials, a normalized density function $\rho(\mathbf{y}) \in [0, 1]$ is defined, which takes the value 1 in regions where the conductive additive material is present, 0.5 in regions where the active material is present, and 0 in void regions. It should be noted that although multiple design variable schemes are commonly employed in optimization methods (Sigmund and Torquato, 1997; Hvejsel and Lund, 2011), the present method is developed solely for analytical purposes.

Accordingly, such schemes were not incorporated, as their introduction offered no significant advantage in the evaluation of the diffusion coefficient. Following the abovementioned approach, a normalized density function is generated.

Based on an arbitrary random function, we obtain sets of sphere center coordinates $[\tilde{y}_{11,1}, \tilde{y}_{12,1}, \tilde{y}_{13,1}, \dots, \tilde{y}_{1g_1,1}]$ for the conductive additive material and $[\tilde{y}_{21,1}, \tilde{y}_{22,1}, \tilde{y}_{23,1}, \dots, \tilde{y}_{21,g_2}, \tilde{y}_{22,g_2}, \tilde{y}_{23,g_2}]$ for the active material with values between 0 and 1. Here, g_1 and g_2 are the numbers of spheres to be generated for conductive additive and active materials, respectively. The diameters of the spheres for conductive additive material R_{s1} and active material R_{s2} are generated from normal distributions on a logarithmic scale, with means of $\log_{10} R_{s1}$ and $\log_{10} R_{s2}$, respectively, and a standard deviation of s . Similarly to a single-material case, R_{s1} and R_{s2} represent the mean ratios of the actual material diameters of the active material and conductive additive material to the size of the unit cell L , respectively. Based on these datasets, a density function $\bar{\rho}$ that takes a value of 1 inside the sphere of conductive additive material and 0.5 inside the sphere of active material is defined as follows:

$$\bar{\rho} = \begin{cases} 1 & \left(\text{if } (y_1 - \tilde{y}_{1,m})^2 + (y_2 - \tilde{y}_{2,m})^2 + (y_3 - \tilde{y}_{3,m})^2 > \left(\frac{R_{s2,m}}{2} \right)^2 \right. \\ & \quad \left. \cap (y_1 - \tilde{y}_{1,n})^2 + (y_2 - \tilde{y}_{2,n})^2 + (y_3 - \tilde{y}_{3,n})^2 \leq \left(\frac{R_{s1,n}}{2} \right)^2 \right) \\ 0.5 & \left(\text{if } (y_1 - \tilde{y}_{1,m})^2 + (y_2 - \tilde{y}_{2,m})^2 + (y_3 - \tilde{y}_{3,m})^2 \leq \left(\frac{R_{s2,m}}{2} \right)^2 \right) \\ 0 & \text{(others),} \end{cases} \quad (4.63)$$

where n is $(1, 2, \dots, g_1)$ and m is $(1, 2, \dots, g_2)$, respectively. As mentioned before, $\bar{\rho}$ does not have a smooth isosurface in the discretized space for numerical analysis. Therefore, $\bar{\rho}$ is smoothed using a Helmholtz filter as defined in Eqs. (4.60) and (4.61). After that, $\bar{\rho}$ is binarized using the following continuous piece-wise smoothed Heaviside projection:

$$\rho = \frac{1}{2} \left\{ \frac{\tanh\{\beta_Y(2\hat{\rho} - 0.5)\} + \tanh(0.5\beta_Y)}{2 \tanh(0.5\beta_Y)} \right\} + \frac{1}{2} \left\{ \frac{\tanh\{\beta_Y(2\hat{\rho} - 1.5)\} + \tanh(0.5\beta_Y)}{2 \tanh(0.5\beta_Y)} \right\}. \quad (4.64)$$

Figure 4.5 illustrates several material distributions with two types of materials generated using the aforementioned method.

4.3.2 Estimation of normalized transport coefficient and surface area

This section describes a method for obtaining the transport coefficients in three directions and the surface area of materials for the given material distribution value ρ obtained in the previous section. In order to obtain the transport coefficients, this paper solves the dimensionless diffusion equations. In other words, the homogeneous Dirichlet boundary condition is given for the direction in which the transport coefficient is to be obtained, and the flux in that direction is calculated. The surfaces in the other directions are applied with the zero-Neumann boundary condition. To

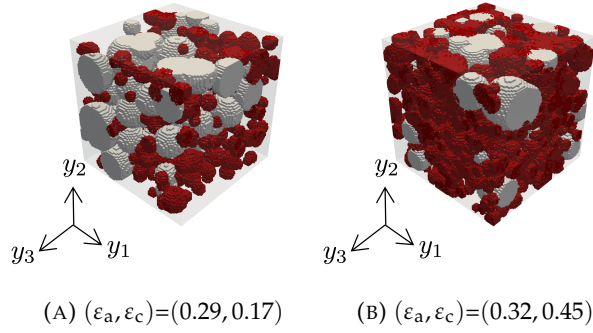


FIGURE 4.5: Randomly generated material distributions in a unit cell for two material types: $R_{s1} = 0.1$ (red) and $R_{s2} = 0.2$ (gray) with a standard deviation $s = 0.10$

evaluate the transport coefficient in the y_1 -direction, we solve the following diffusion equations:

$$\mathbf{q}_1 = -K \nabla_Y C_1, \quad (4.65)$$

$$\nabla_Y \cdot \mathbf{q}_1 = 0 \quad \text{in } Y, \quad (4.66)$$

$$C_1 = 0 \quad \text{on } \Gamma_1, \quad (4.67)$$

$$C_1 = 1 \quad \text{on } \Gamma_2, \quad (4.68)$$

$$\mathbf{q}_1 \cdot \mathbf{n} = 0 \quad \text{on } \Gamma_{3,4,5,6}, \quad (4.69)$$

where C_1 is the dimensionless scalar field and K is the dimensionless transport coefficient. As for the material distribution of single type of material, K is defined as 1 where there is material and a smaller value $K_{\min}$ where there is no material as follows:

$$K = \rho + (1 - \rho)K_{\min}. \quad (4.70)$$

Whereas, as for the material distribution of both active material and conductive additive material, K is defined as $K_{\max}$ where there is conductive additive material, 1 where active material exists, and a smaller value $K_{\min}$ where there is no material as follows:

$$K = (1 - K_{\min}) \left\{ \frac{\tanh\{\beta_Y(2\hat{\rho} - 0.5)\} + \tanh(0.5\beta_Y)}{2 \tanh(0.5\beta_Y)} \right\} + K_{\min} \\ + (K_{\max} - 1) \left\{ \frac{\tanh\{\beta_Y(2\hat{\rho} - 1.5)\} + \tanh(0.5\beta_Y)}{2 \tanh(0.5\beta_Y)} \right\}, \quad (4.71)$$

where $K_{\max}$ is the electronic conductivity of the conductive additive material normalized by that of the active material as follows:

$$K_{\max} = \frac{\sigma_c}{\sigma_a}, \quad (4.72)$$

where σ_c is the electronic conductivity of the conductive additive material. The flux of C_1 , Q_1 , in the y_1 direction is then calculated based on the following equation:

$$W_1 = Q_1 = \int_{\Gamma_1} \mathbf{q}_1 \cdot \mathbf{n} \, d\Gamma, \quad (4.73)$$

where the dimensionless effective transport coefficient W_1 in the y_1 direction coincides with Q_1 since the size of the unit cell is 1 and the difference between the values of the Dirichlet conditions at boundaries Γ_1 and Γ_2 is set to 1.

In the same way, the dimensionless effective transport coefficients, W_2 and W_3 , in the y_2 and y_3 directions, respectively, are obtained. The following dimensionless diffusion equations for the dimensionless scalar fields, C_2 and C_3 , are solved:

$$\mathbf{q}_2 = -K \nabla_Y C_2, \quad (4.74)$$

$$\nabla_Y \cdot \mathbf{q}_2 = 0 \quad \text{in } Y, \quad (4.75)$$

$$C_2 = 0 \quad \text{on } \Gamma_3, \quad (4.76)$$

$$C_2 = 1 \quad \text{on } \Gamma_4, \quad (4.77)$$

$$\mathbf{q}_2 \cdot \mathbf{n} = 0 \quad \text{on } \Gamma_{1,2,5,6}, \quad (4.78)$$

$$\mathbf{q}_3 = -K \nabla_Y C_3, \quad (4.79)$$

$$\nabla_Y \cdot \mathbf{q}_3 = 0 \quad \text{in } Y, \quad (4.80)$$

$$C_3 = 0 \quad \text{on } \Gamma_5, \quad (4.81)$$

$$C_3 = 1 \quad \text{on } \Gamma_6, \quad (4.82)$$

$$\mathbf{q}_3 \cdot \mathbf{n} = 0 \quad \text{on } \Gamma_{1,2,3,4}. \quad (4.83)$$

Then, the flux of C_2 and C_3 in the y_2 and y_3 directions, Q_2 and Q_3 , respectively, are evaluated by the following equations, respectively:

$$W_2 = Q_2 = \int_{\Gamma_3} \mathbf{q}_2 \cdot \mathbf{n} \, d\Gamma, \quad (4.84)$$

$$W_3 = Q_3 = \int_{\Gamma_5} \mathbf{q}_3 \cdot \mathbf{n} \, d\Gamma. \quad (4.85)$$

Similarly, Q_2 and W_2 , and Q_3 and W_3 coincide. Based on the transport coefficients obtained from these analyses, isotropic transport coefficient $\tilde{H}$ is derived using the L_2 norm as follows:

$$\tilde{H}(\rho(\mathbf{y}; \varepsilon)) = \frac{\sqrt{W_1^2 + W_2^2 + W_3^2}}{\sqrt{3}}, \quad (4.86)$$

Next, the surface area of randomly generated material distribution in the unit cell is evaluated using the following equation, which is one of the bases of the Hamilton-Jacobi equation (Osher

and Sethian, 1988):

$$\tilde{A}(\rho(\mathbf{y}; \varepsilon)) = \int_Y |\nabla \rho| \, dY. \quad (4.87)$$

Accuracy in the data acquisition phase, a prerequisite for optimization, is of critical importance. For a more precise calculation of the surface area, methods such as a body-fitted mesh (Dapogny et al., 2014) or the Extended Finite Element Method (Moës et al., 1999) are desirable. However, considering the computational cost required to evaluate a large number of data points, the proposed method employs a surface area calculation approach based on the density method.

4.3.3 Algorithm for obtaining interpolation functions

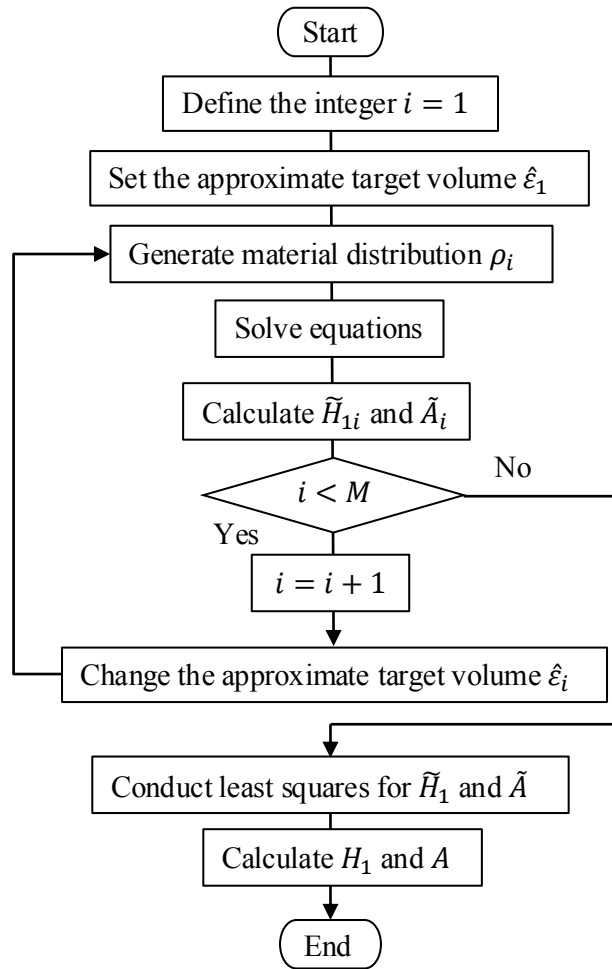


FIGURE 4.6: Algorithm for making the material interpolation function H_1 and A

This section shows the algorithm details for obtaining the interpolation functions, $H_1(\varepsilon)$, $H_2(\varepsilon_a, \varepsilon_c)$ and $A(\varepsilon)$, which depend on the volume fraction of the material. First, we explain the algorithm for finding $H_1(\varepsilon)$ and $A(\varepsilon)$ which depend on only one volume fraction. Figure 4.6 shows the flowchart

of the proposed algorithm for obtaining $H_1(\varepsilon)$ and $A(\varepsilon)$. Initially, the integer i is defined as 1 to count the number of loops. Then, a target value for the volume fraction, $\hat{\varepsilon}_i$, which is used when the material distribution is created, is set as 0. Next, the spheres' center coordinates are defined randomly so that the material volume approaches the target value. After that, the actual volume of the generated material distribution ε_i is obtained. Note that there is no guarantee that $\hat{\varepsilon}_i$ and ε_i coincide because of the existence of overlaps. Next, filtering and binarization with the smoothed Heaviside function are performed on the generated material distribution as shown in Eq. (4.60) – (4.62), Eqs. (4.65)–(4.70), and Eqs. (4.74)–(4.83) are then solved to obtain the effective transport coefficient $\tilde{H}_{1i}$ and surface area $\tilde{A}_i$ from Eqs. (4.86) and (4.87), respectively. If the number of loops i is less than M , the target value of the volume fraction $\hat{\varepsilon}_i$ is incrementally increased up to 1, and the same procedure is conducted.

At the end of M loops, a dataset of volume fractions $\varepsilon_i (i = 1, \dots, M)$, transport coefficients $\tilde{H}_{1i} (i = 1, \dots, M)$, and surface areas $\tilde{A}_i (i = 1, \dots, M)$ for M varieties of material distributions $\rho_i (i = 1, \dots, M)$ can be obtained. Here, the functions $\tilde{H}_1$ and $\tilde{A}$ are interpolated by a U -dimensional polynomial of the volume fractions, as shown in the following equations:

$$\tilde{H}_1 = \sum_{m=0}^U h_m \varepsilon^m, \quad (4.88)$$

$$\tilde{A} = \sum_{m=0}^U w_m \varepsilon^m, \quad (4.89)$$

where h_m and w_m are the coefficients of the respective polynomial equations. Then, the least-squares method is applied to each equation using the acquired dataset to obtain $\tilde{H}_1$ and $\tilde{A}$.

Next, the algorithm for finding $H_2(\varepsilon_a, \varepsilon_c)$ which depends on the volume fractions of both active material and conductive additive material. Initially, the integer i is defined as 1 to count the number of loops. Then, target values for the volume fraction of active material and conductive additive material, $\hat{\varepsilon}_{a,i}$ and $\hat{\varepsilon}_{c,i}$, are set. Next, the spheres' center coordinates are defined randomly so that the material volume approaches the target values. After that, the actual volumes of the generated material distribution, $\varepsilon_{a,i}$ and $\varepsilon_{c,i}$, are obtained. Next, filtering and binarization with the continuous piece-wise smoothed Heaviside function are performed on the generated material distribution as shown in Eqs. (4.60)–(4.61), Eq. (4.64), Eqs. (4.65)–(4.69), Eq. (4.71), and Eqs. (4.74)–(4.83) are then solved to obtain the effective transport coefficient $\tilde{H}_{2i}$ from Eq. (4.86). If the number of loops i is less than M , the target values of the volume fractions, $\hat{\varepsilon}_{a,i}$ and $\hat{\varepsilon}_{c,i}$, are updated, and the same procedure is conducted.

At the end of M loops, a dataset of volume fractions, $\varepsilon_{a,i}$ and $\varepsilon_{c,i} (i = 1, \dots, M)$, and transport coefficients $\tilde{H}_{2i} (i = 1, \dots, M)$ for M varieties of material distributions $\rho_i (i = 1, \dots, M)$ can be obtained. Here, the function $\tilde{H}_2$ is interpolated by a U_a -dimensional polynomial for ε_a and a U_c -dimensional

polynomial for ε_c , as shown in the following equation:

$$\tilde{H}_2(\varepsilon_a, \varepsilon_c) = \sum_{m=0}^{U_a} \sum_{j=0}^{U_c} h_{a,m} h_{c,j} \varepsilon_a^m \varepsilon_c^j, \quad (4.90)$$

where $h_{a,m}$ and $h_{c,j}$ are the coefficients of the polynomial equations. At the end, the least squares method is applied to the equation using the acquired dataset to obtain $\tilde{H}_2$.

Here, the functions $\tilde{H}_1$ and $\tilde{H}_2$ obtained by this method have a fatal drawback in optimization problems. That is, it does not guarantee to have a monotonic increase property and positive value. The interpolation function obtained from pure least squares may take negative values and may oscillate slightly. These characteristics are different from the physical interpretation and have a harmful impact on the search for an optimization solution that is updated based on the gradient of variables. Therefore, we consider the monotonic increase and positive constraints when the least squares for $\tilde{H}_1$ and $\tilde{H}_2$ are conducted. In other words, we impose the following constraints:

$$\sum_{m=0}^U h_m \varepsilon_i^m > 0 \quad (i = 1, \dots, M), \quad (4.91)$$

$$\sum_{m=1}^U m h_m \varepsilon_i^{m-1} > 0 \quad (i = 1, \dots, M), \quad (4.92)$$

for $\tilde{H}_1$, and

$$\sum_{m=0}^{U_a} \sum_{j=0}^{U_c} h_{a,m} h_{c,j} \varepsilon_{a,i}^m \varepsilon_{c,i}^j > 0 \quad (i = 1, \dots, M), \quad (4.93)$$

$$\sum_{m=1}^{U_a} \sum_{j=0}^{U_c} m h_{a,m} h_{c,j} \varepsilon_{a,i}^{m-1} \varepsilon_{c,i}^j > 0 \quad (i = 1, \dots, M), \quad (4.94)$$

$$\sum_{m=0}^{U_a} \sum_{j=1}^{U_c} j h_{a,m} h_{c,j} \varepsilon_{a,i}^m \varepsilon_{c,i}^{j-1} > 0 \quad (i = 1, \dots, M), \quad (4.95)$$

for $\tilde{H}_2$. The implemented code for the proposed interpolation method is presented in Appendix B.2.

Moreover, the presence of Ersatz material should be considered to stabilize the convergence of the numerical calculation during the optimization process as follows:

$$H_1 = \tilde{H}_1(\varepsilon) + H_{1,\min}, \quad (4.96)$$

$$H_2 = \tilde{H}_2(\varepsilon_a, \varepsilon_c) + H_{2,\min}, \quad (4.97)$$

where $H_{1,\min}$ and $H_{2,\min}$ are the given small values. Similarly, since $\tilde{A}$ has a minimal value around zero, $A_{\min}$ is also added to stabilize the convergence of the numerical calculation as follows:

$$A = \tilde{A}(\varepsilon) + A_{\min}. \quad (4.98)$$

Note that only the positive value constraint is considered when $\tilde{A}(\varepsilon)$ is derived using the least squares method, because there is no physical basis requiring $\tilde{A}(\varepsilon)$ to be monotonically increasing.

4.4 Numerical implementations

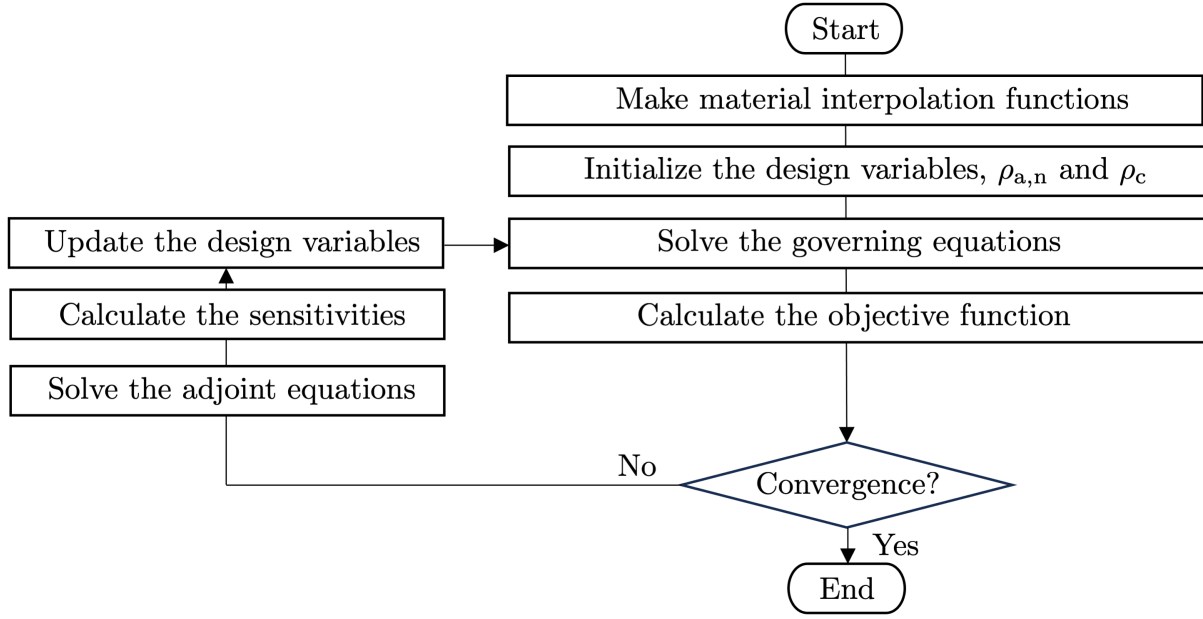


FIGURE 4.7: Algorithm for the optimization

This section describes the numerical implementation details for the optimization problem formulated in Section 4.2.3. Figure 4.7 shows the optimization algorithm used in this paper. First, the material interpolation function values, H_1 , H_2 and A , are computed using the method explained in Section 4.3. Then, initial configurations of the design variables, $\rho_{a,n}$ and ρ_c , are given. Next, the governing equations Eqs. (4.3)–(4.18) are solved based on the design variables from $t = 0$ until the termination condition Eq. (4.21) is satisfied. After that, the objective function J is evaluated. Then, the termination criterion is checked using the following conditions:

$$\left\| \frac{J_{r+1} - J_r}{J_r} \right\| \leq \omega, \quad (4.99)$$

$$\left\| \frac{N_{1,r+1} - N_{1,r}}{N_{1,r}} \right\| \leq \omega, \quad (4.100)$$

$$\left\| \frac{N_{2,r+1} - N_{2,r}}{N_{2,r}} \right\| \leq \omega, \quad (4.101)$$

where the index r is the number of the optimization procedure, and ω is the given convergence criterion. If Eqs. (4.99)–(4.101) are satisfied five consecutive times, the convergence is considered to be achieved, and the optimization process is terminated. On the other hand, if the equation

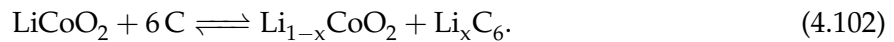
is not satisfied, the adjoint equations Eqs. (4.40)–(4.55) are solved. Then, the sensitivities, J'_1 and J'_2 , are calculated based on the state and adjoint variables. Finally, the design variables are updated using the Method of Moving Asymptotes (Svanberg, 1987), and the governing equations are solved again.

4.5 Numerical examples

In this section, several numerical examples are presented to confirm the availability of the proposed method. Section 4.5.1 presents the parameter settings used in this numerical example. Section 4.5.2 presents detailed numerical examples and discusses the physical meanings of the obtained material interpolation functions, H_1 , H_2m and A , based on the method described in Section 4.3. Finally, Section 4.5.3 shows the optimal solution obtained by conducting the proposed optimization method, i.e., the optimal distribution of the volume fractions of each three materials existing in the composite anode. In this paper, for the microstructural analysis, FreeFEM++ is used, while the optimization was performed using COMSOL Multiphysics Simulations with Matlab®, a solver with a functional standing for battery analysis.

4.5.1 Numerical settings

Table 4.2 shows the values of the constants used in the numerical examples. This paper used lithium cobalt oxide for the cathode and graphite carbon for the anode. The equilibrium potential equations, U_n and U_p , were based on the material data provided by COMSOL Multiphysics (Multiphysics, 1998). It is assumed that electrochemical reactions take place inside the battery based on the following chemical reaction:



Here, the right-to-left reaction is the discharge reaction, in which Li is deintercalated from the anode region. In contrast, the left-to-right reaction is the charge reaction, in which Li is intercalated into the anode region.

4.5.2 Material interpolation function profile

Figure 4.8 shows the discretization mesh used for the microstructural analysis. The tetrahedral elements with size of $1/80$ were utilized. The dimensionless numbers R_s , R_{s1} , and R_{s2} was set to 0.2, 0.1, and 0.2, respectively, where R_s and R_{s2} are the mean ratio of the actual material diameter of the electrolyte and active materials to the size of the unit cell L to be considered, and R_{s1} is the mean ratio of the actual material diameter of the conductive additive material to the size of

TABLE 4.2: Parameter settings

Parameter	Value	Unit
L_n	85.0	μm
L_e	45.0	μm
L_p	65.0	μm
L_h	100	μm
L	10	μm
A_g	1	m
$\varepsilon_{a,p}$	0.44	–
$\varepsilon_{e,e}$	0.60	–
$\varepsilon_{e,p}$	0.40	–
$\overline{\varepsilon_{a,n}}$	0.55	–
$\frac{\varepsilon_{a,n}}{\varepsilon_c}$	0	–
$\frac{\varepsilon_c}{\varepsilon_c}$	0.40	–
$\frac{\varepsilon_c}{\varepsilon_c}$	0	–
C_{rate}	1	–
$\text{SOC}_{n,\text{max}}$	0.98	–
$\text{SOC}_{n,\text{min}}$	0	–
$\text{SOC}_{p,\text{max}}$	1	–
$\text{SOC}_{p,\text{min}}$	0.43	–
$c_{n,\text{max}}$	31507	mol/m^3
$c_{p,\text{max}}$	56250	mol/m^3
F	96485.3	C/mol
R	8.3143	J/(mol · K)
T	298	K
$V_{\text{terminate}}$	2.7	V
ω	1.0×10^{-3}	–
$i_{0,n}$	36	A/m ²
$i_{0,p}$	26	A/m ²
D_n	2.09×10^{-15}	m ² /s
D_p	5.00×10^{-13}	m ² /s
σ_n	1.00×10^{-1}	S/m
σ_p	1.13×10^{-2}	mS/cm
σ_c	100	S/m
κ	2.00×10^{-2}	S/m
M	(250, 420)	–
β_Y	30	–
$H_{1,\text{min}}$	0.001	–
$H_{2,\text{min}}$	0.001	–
A_{min}	1	–
K_{min}	0.001	–
K_{max}	1000	–
$\overline{V}_a$	0.4	–
$\overline{V}_c$	0.35	–

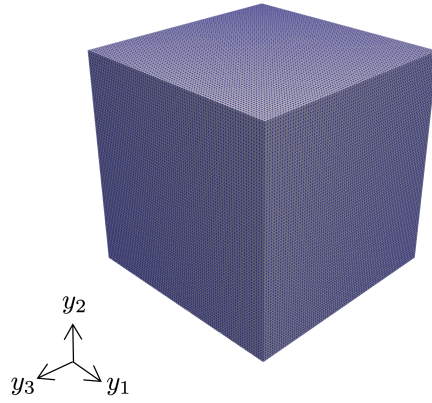


FIGURE 4.8: Mesh for the unit cell

the unit cell. Here, as an example in this study, we obtained the characteristics of the physical properties observed in a unit cell with a size L of 10 [μm], assuming that the diameters of the electrolyte and active materials were 2 [μm] and that of the conductive additive was 1 [μm]. The number of microstructure analysis loops M was set to 250 for H_1 and A , and 420 for H_2 . The ratio of the electronic conductivity of the conductive additive material to that of the active material, K_{max} , was set to 1000.

First, we validate the random generation process. In this study, uncertainty is introduced into both position coordinates and the diameters of the spheres. For the position coordinates, a uniform distribution is generated using the "srandomdev" function provided in FreeFEM++. To generate the diameters, which follow a normal distribution, we employ random sampling based on the Box-Muller method (Box and Muller, 1958). Specifically, a standard normal distribution is obtained from uniformly distributed random variables generated by the "srandomdev" function. Figure 4.9 presents the histograms and probability density functions of log-normal distributions, where 100 data points are sampled for the following implemented three probability distributions. The distributions have a standard deviation of 0.1, and the means of the base-10 logarithm of the diameters $\log_{10} R_s$ are set to $\log_{10} 0.2$, $\log_{10} 0.1$, and $\log_{10} 0.04$, respectively. Here, the probability density functions of log-normal distributions with a mean of $\log_{10} R_s$ and a standard deviation s are denoted as follows:

$$f(R)(R_s, s) = \frac{1}{R \log_e 10 s \sqrt{2\pi}} \exp\left(-\frac{(\log_{10} R - \log_{10} R_s)^2}{2s^2}\right). \quad (4.103)$$

As Fig. 4.9 shows, the histograms obtained by sampling 100 data points for each log-normal distribution with the given mean align with the corresponding probability density functions. This demonstrates the validity of our implemented random generation process.

Next, Fig. 4.10 presents the obtained data points and the constructed interpolation functions,

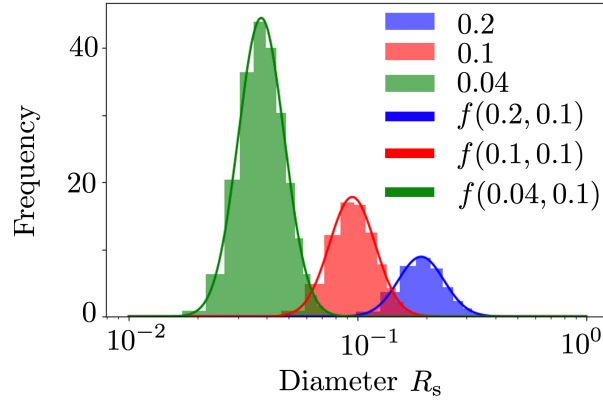
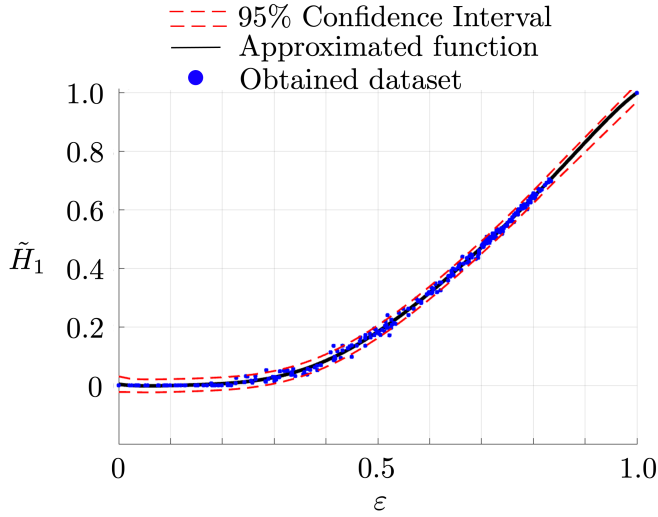


FIGURE 4.9: Histograms of the sampled 100 points and probability density functions $f(R_s, s)$ of log-normal distributions

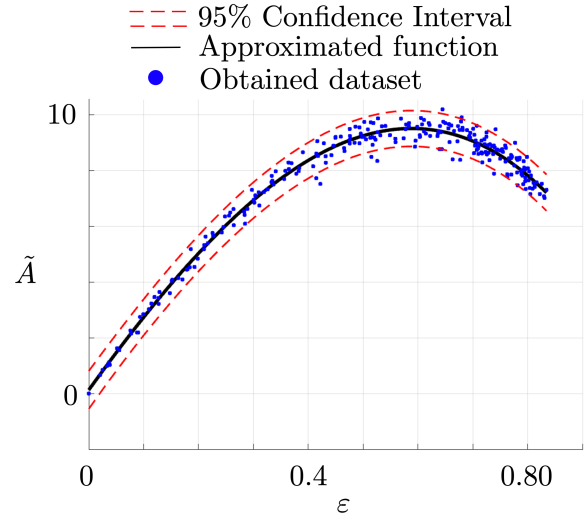
$\tilde{H}_1$, $\tilde{H}_2$, and $\tilde{A}$, as a function of the volume fraction with R_s of 0.2. Especially, in Figs. 4.10a and b, the 95% confidence intervals for each interpolation function are depicted. As Fig. 4.10 shows, monotonically increasing functions $\tilde{H}_1$ and $\tilde{H}_2$, and an upward convex function $\tilde{A}$ interpolate the acquired datasets well. Comparing the widths of the confidence intervals for $\tilde{H}_1$ and $\tilde{A}$, $\tilde{A}$ exhibits a larger interval, indicating greater variability in surface area. This is because, even if the volume fraction is the same, there can be many different ways to distribute the materials, and each of those patterns can result in a different surface area. It should be noted that an additional data-point of $\tilde{H}_1 = 1$ at $\varepsilon = 1$ was considered when creating the interpolation function $\tilde{H}_1$, since the transport coefficient at $\varepsilon = 1$ is the same as that of the bulk material.

Next, we examine the variation of the obtained data in more detail with respect to the interpolation function. Figure 4.11 presents the absolute error (vertical axis) between the obtained data points and the interpolating function derived using the least-squares method, along with the relative error (color), where the interpolating function serves as the reference. Figure 4.11a shows that variation is extremely small for volume fractions of approximately 0.2 or less. This occurs because, in this range, the material does not form a continuous transportation path across the unit cell. Meanwhile, for volume fractions slightly above 0.2, some data points exhibit a relative error of 200%. As shown in Fig. 4.10a, this corresponds to the transition between the presence and absence of a transportation path at a volume fraction of 0.2, where the diffusion coefficient begins to increase. Consequently, this region is highly sensitive to variations in material distribution. On the other hand, according to Fig. 4.11b, the magnitude of the absolute error in surface area is approximately 20 times greater than that of the diffusion coefficient. However, the relative error analysis indicates a maximum relative error of 12%, suggesting that the overall variation is small. This implies that the probability distribution affects surface area similarly across all volume fractions. These findings align with physical considerations, supporting the validity of our method.

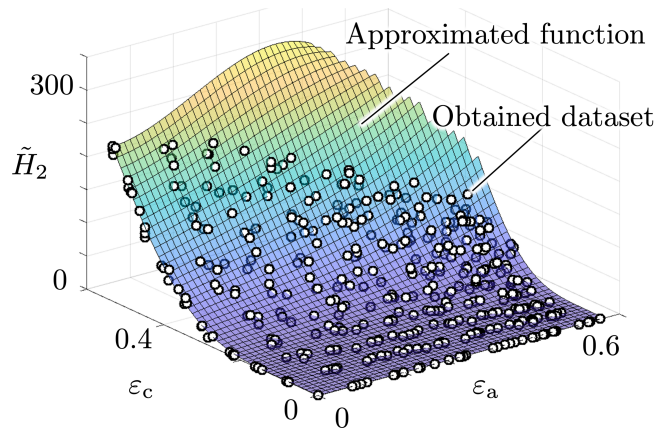
Next, we consider the effect of the orders of the interpolation function shown in Eqs. (4.88)–(4.90) on the function configurations. The order used in the least square is determined by trial



(A) For the dimensionless transport coefficient which depends on single type of material, $U = 7$



(B) For the dimensionless surface area, $U = 3$



(C) For the dimensionless transport coefficient which depends on two types of materials, $U_a = 3$ and $U_c = 5$

FIGURE 4.10: Comparison of data-points from microstructure analysis and the approximated material interpolation functions, $\tilde{H}_1$, $\tilde{H}_2$ and $\tilde{A}$ with $R_s = 0.2$ and $s = 0.1$

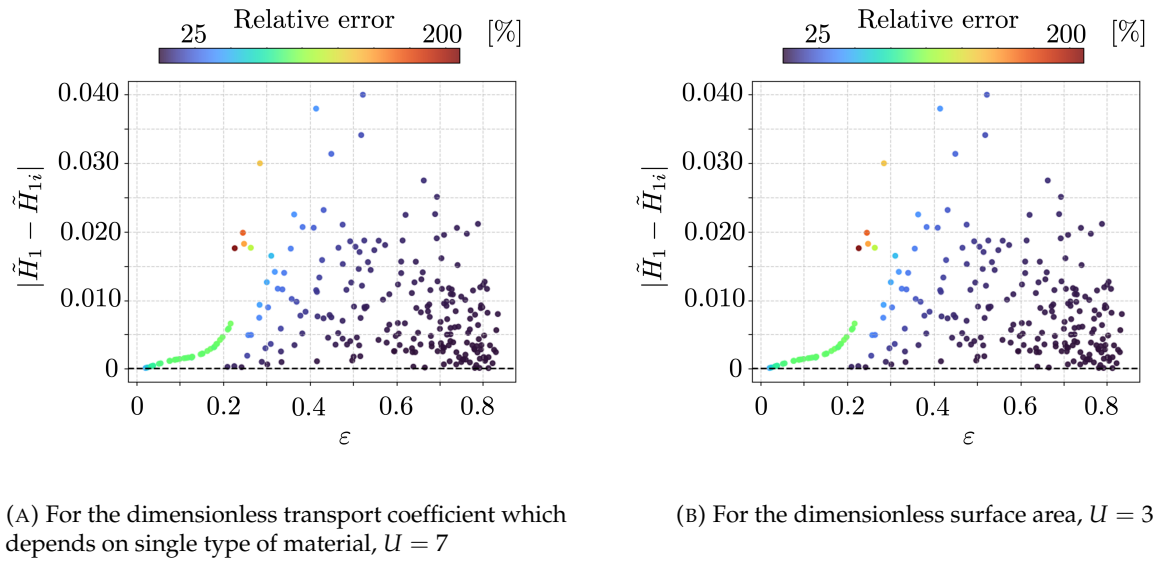


FIGURE 4.11: Visualization of absolute (vertical axis) and relative (color) errors between data points and approximated material interpolation function

TABLE 4.3: The coefficients h for $\tilde{H}_1$ obtained by least squares for different values U and the values of AIC

U	h_0	h_1	h_2	h_3	h_4	h_5	h_6	h_7	h_8	AIC
5	1.10×10^{-12}	0.131	-1.44	5.81	-4.42	0.908	-	-	-	-2255
6	6.98×10^{-10}	3.86×10^{-2}	-0.411	0.751	7.20	-11.2	4.65	-	-	-2269
7	3.33×10^{-11}	4.33×10^{-9}	0.630	-7.95	38.5	-65.9	50.4	-14.7	-	-2271
8	1.3×10^{-9}	1.2×10^{-2}	-0.35	4.5	-26	101	-181	148	-45	-2270

TABLE 4.4: The coefficients w for $\tilde{A}$ obtained by least squares for different values U and the values of AIC

U	w_0	w_1	w_2	w_3	w_4	w_5	AIC
2	-0.400	33.8	-29.2	-	-	-	-524.6
3	0.130	27.4	-11.7	-13.1	-	-	-560.8
4	-0.0187	30.0	-24.8	-9.86	-13.1	-	-560.7
5	0.134	25.8	7.59	-87.4	112	-57.6	-560.3

TABLE 4.5: AIC values for varying orders of least squares, U_a and U_c

$U_a \backslash U_c$	4	5	6
2	1396	1338	1331
3	1379	1328	1330
4	1385	1339	1342

and error approach, and there is no existing method to determine it uniquely. Therefore, in this numerical example, Akaike information criterion(AIC) was utilized as a criterion.

Tables 4.3 and 4.4 present the coefficients and AIC values for $\tilde{H}_1$ and $\tilde{A}$ obtained through least squares fitting for various orders U . For $\tilde{H}_1$, the seventh-order function yielded the smallest AIC value, while for $\tilde{A}$, the smallest AIC value was observed at $U = 3$. Accordingly, the orders of $\tilde{H}_1$ and $\tilde{A}$ were set to 7 and 3, respectively. Meanwhile, Table 4.5 presents the AIC values for $\tilde{H}_2$ obtained through least squares fitting with various orders of U_a and U_c . The combination of $U_a = 3$ and $U_c = 5$ yielded the smallest AIC value. Accordingly, this study adopts this pair for the least squares fitting of $\tilde{H}_2$, resulting in the following equation:

$$\begin{aligned} \tilde{H}_2(\varepsilon_a, \varepsilon_c) = & 1.30 \times 10^{-12} + 6.75 \times 10^{-11} \varepsilon_c^1 + 279 \varepsilon_c^2 - 4150 \varepsilon_c^3 + 18900 \varepsilon_c^4 - 17400 \varepsilon_c^5 \\ & + \varepsilon_a(9.48 \times 10^{-2} + 23.0 \varepsilon_c^1 - 450 \varepsilon_c^2 - 128 \varepsilon_c^3 + 29900 \varepsilon_c^4 - 52300 \varepsilon_c^5) \\ & + \varepsilon_a^2(-0.685 + 106 \varepsilon_c^1 - 3440 \varepsilon_c^2 + 40700 \varepsilon_c^3 - 198000 \varepsilon_c^4 + 288000 \varepsilon_c^5) \\ & + \varepsilon_a^3(1.58 - 164 \varepsilon_c^1 + 5250 \varepsilon_c^2 - 64200 \varepsilon_c^3 + 362000 \varepsilon_c^4 - 554000 \varepsilon_c^5). \end{aligned} \quad (4.104)$$

Figure 4.12 illustrates the plots of the proposed function H_1 with R_s of 0.2, 0.1, and 0.04 and the SIMP method with penalty parameters $p = 1.5, 2$, and 3 as a function of the volume fraction. It should be noted that the preceding discussion is based on interpolating functions obtained for $R_s = 0.2$. To examine how the interpolating functions change with varying R_s , the same procedure was applied to generate additional interpolating functions for $R_s = 0.1$ and 0.04. As shown in Fig. 4.12, the profile of the approximated interpolation function varies with the particle mean diameter R_s . As R_s decreases, the interpolation function approaches the Bruggeman model ($p = 1.5$). This suggests that the interpolation function must be appropriately developed based on the utilized material to conduct the optimization under precise conditions.

Comparing the interpolation function obtained for the smallest $R_s = 0.04$ with that based on the Bruggeman model, the interpolation function exhibits lower values at small volume fractions. This aligns with previous findings that the Bruggeman model tends to overestimate in this regime (Tjaden et al., 2016). Thus, the proposed method for constructing interpolation functions is consistent with experimental observations.

Furthermore, Fig. 4.12 indicate that the behavior of the proposed function H_1 with $R_s = 0.2$ differs from that of the SIMP method. For small volume fractions, H_1 closely resembles the distribution with a penalty parameter of $p = 3$, whereas for larger volume fractions, it approaches the configuration with $p = 2$. In the context of this study, where intermediate material properties are incorporated into the optimal solution, these differences significantly influence the optimized configuration, as discussed in Section 4.5.3. Consequently, the material interpolation function derived through the physical interpretation presented here plays a critical role in optimization problems for battery designs.

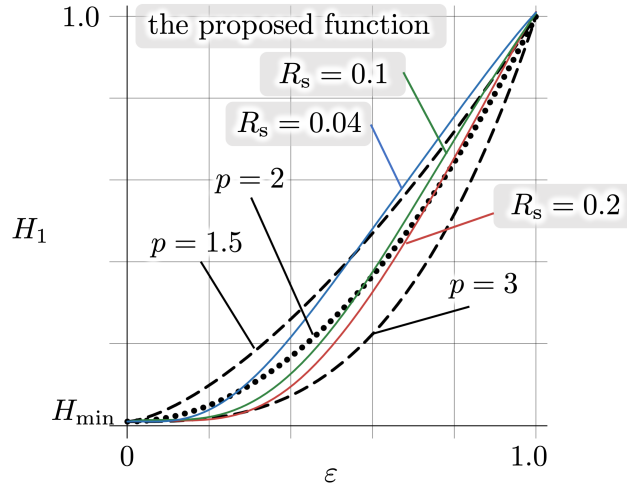


FIGURE 4.12: Plots of the approximated material interpolation function H_1 for the dimensionless transport coefficient with $R_s = 0.2, 0.1$, and 0.04 and $s = 0.1$, and SIMP method with $p = 1.5, 2$, and 3 as a function of volume fraction

Here, the proposed method for generating material distributions is validated through comparison with experimental data (Ebner et al., 2014). In a previous study, the tortuosity p_t described by the following equation was derived using the least-squares method based on several actual material distributions,

$$H_1 = \varepsilon^{(1+p_t)}. \quad (4.105)$$

The optimal tortuosity p_t is also determined by applying the least-squares method to the data points obtained through the proposed method, using the same functional form, Eq. (4.105). It should be mentioned that the previous studies refer to the particle diameter but do not specify the unit cell size, indicating that the conditions are not strictly consistent. Figure 4.13 presents the data points obtained when $R_s = 0.04$ and $s = 0.1$, along with the profile of Eq. (4.105) fitted to these points. The optimal value of p_t was 0.5153. This result is generally consistent with experimental data for NMC composed of spherical particles, as shown in Table 1 of the referenced study (Ebner et al., 2014). Furthermore, the obtained value is also close to that predicted by the well-known Bruggeman model, $p_t = 0.5$. These comparisons suggest that the material generation method and the diffusion coefficient evaluation method used in the proposed approach are reasonably consistent with the experimental data.

Finally, the computational cost taken for this numerical example is mentioned. The numerical examples were performed on a Mac Studio equipped with an Apple M1 Ultra chip and 64 GB of memory. The average computation time for the 3D analysis of a given material distribution was approximately 87.5 seconds using 10 parallel processes with PETSc. On the other hand, the computational cost for generating material distributions depends on the number of spheres. For instance, generating 106 and 290 spheres took 2.66 seconds and 7.18 seconds, respectively.

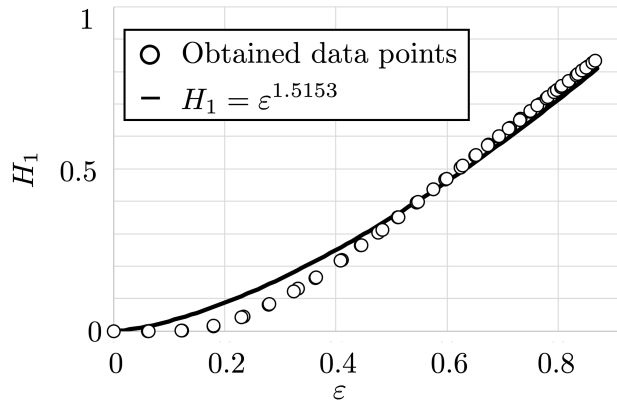


FIGURE 4.13: Plots of the obtained data points with $R_s = 0.04$ and $s = 0.1$, and the function profile corresponding to $p_t = 0.5153$ in Eq. 4.105

4.5.3 Optimization results

In this section, the optimized configurations of the material volume fraction obtained by the proposed optimization method are shown to confirm the effectiveness and characteristics of the proposed method. Section 4.5.3 exhibits the performance of the optimized configuration by comparing the performance with a uniform distribution. Section 4.5.3 shows the importance of proposing the material interpolation functions. Section 4.5.3 demonstrates the generality of the proposed method by presenting the variations in the optimal solution when the conductivities of the conductive additive material and active material are altered.

In these numerical examples, the discretized mesh shown in Fig. 4.14 is utilized, consisting of 6,474 nodes. It should be noted again that, in the following numerical optimization examples, we employ $R_s = 0.2$ with $U = 7$ and $U = 3$ for $\tilde{H}_1$ and $\tilde{A}$, respectively, while for $\tilde{H}_2$, the interpolation function shown in Eq. (4.104) is used.

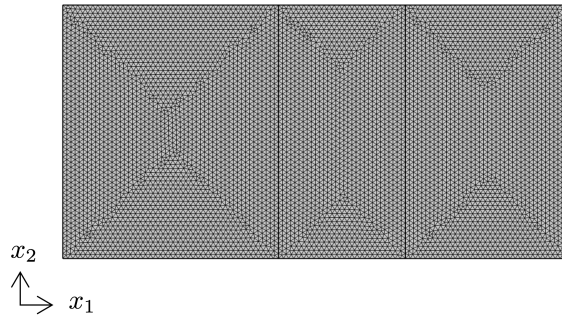


FIGURE 4.14: Mesh for the battery analysis

Performance evaluation of optimized material distribution: Comparison with uniform distribution

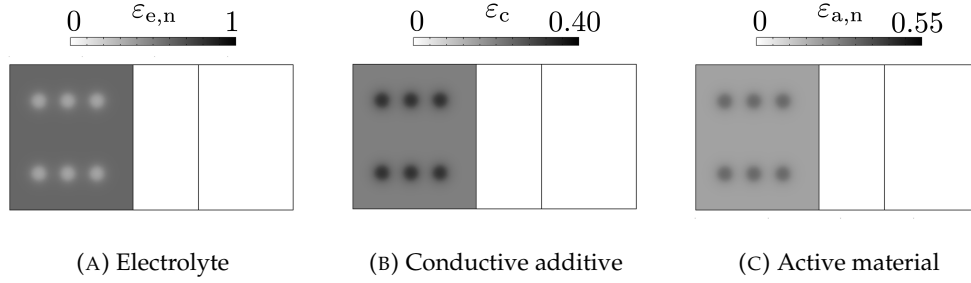


FIGURE 4.15: The initial material distributions for the optimization

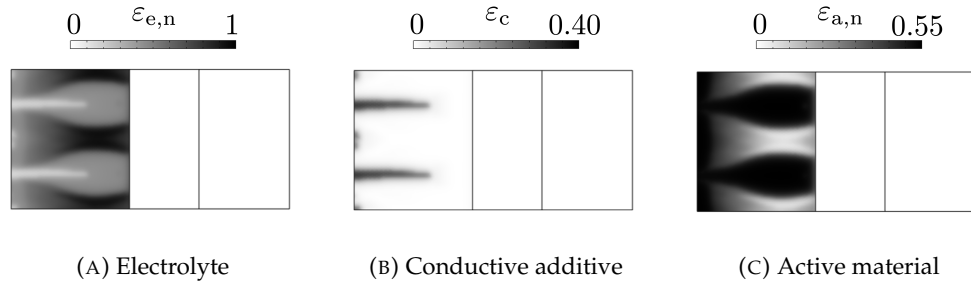


FIGURE 4.16: The obtained material distributions by the proposed method

Figure 4.15 shows the initial configurations of the three materials, and Fig. 4.16 shows the optimized configurations obtained by the proposed method. The average volume fractions of the electrolyte, electrolytic additive, and active material for the obtained results after optimization were 0.571, 0.029, and 0.400, respectively. Figure 4.17 shows the history plots of the objective and constraint function values. These plots demonstrate that each value converges to a certain value. In particular, regarding the constraint functions, $\bar{\varepsilon}_{a,n}$ is active in the optimized results, whereas $\bar{\varepsilon}_c$ is inactive. These results indicate that the volume of the active material tends to increase in order to reduce the objective function value by enhancing the current. On the other hand, the volume of the conductive additive converges to a certain optimal value, which remains below the upper limit $\bar{V}_c$ and serves merely to compensate for the low conductivity of the active material. This observation is reasonable, considering the respective roles of the materials. This optimization calculation took a total of 11,497 seconds on a workstation equipped with an Intel Core i9-13900K processor and 64 GB of memory.

The obtained results exhibit that the electrolyte is distributed mainly on the side close to the electrolyte phase, while the active and conductive additive materials are distributed near the current collector. This tendency is an attempt to reduce the resistance of electron and Li ion conduction during the discharge process and is consistent with the results described in Clausnitzer et al., 2023.

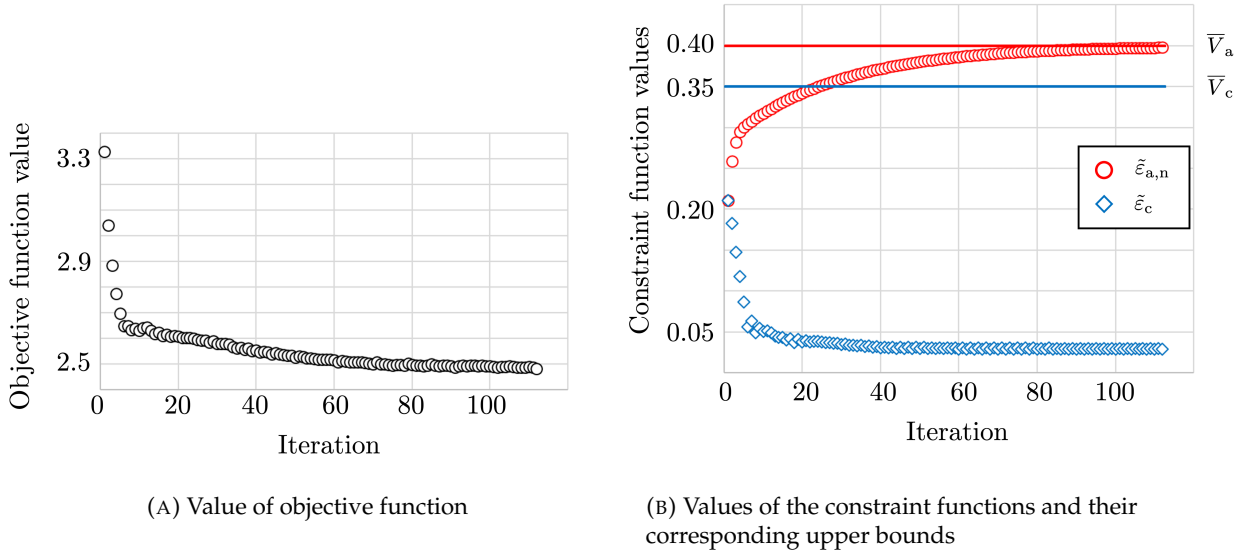


FIGURE 4.17: History plots of objective and constraint values during the optimization iterations

The active material is distributed in a wavy pattern. This distribution attempts to supply Li evenly in the composite anode by arranging the active material, which is responsible for holding Li, in a wave-like form. Such a material distribution design was realized through the high degree of freedom offered by the TO method.

Next, the performance of the obtained configuration is demonstrated by comparing it to a commonly used homogeneously mixed battery. Among them, we consider two types: one with a rectangular composite anode area and the other with a 'parallel' type (Lin et al., 2024), where these configurations are named 'rectangular-type uniform material distribution' and 'parallel-type uniform material distribution', respectively, in this paper. Figure 4.18 shows the battery configuration of each type of material distribution. To ensure consistent battery conditions, including the theoretical capacity and the surface area between active material and electrolyte material, the volume fraction of each material and the size of each domain are set so that the theoretical capacity and the total surface area in the composite anode are equal to the obtained optimal solution. The objective functions of the optimized structure and the rectangular and parallel-type uniform distributions were 2.47, 2.82 and 2.52, respectively. This result indicates that the proposed method reduces over-voltage by 12.4% and 1.98% compared to the rectangular and parallel-type uniform distributions, respectively, under the condition that the theoretical capacities are the same for each case. Furthermore, the SOC of the anode at the termination time for the optimized structure and the uniform material distributions were 0.149, 0.202 and 0.157, respectively. This result demonstrates that the proposed TO method enhances the capacity by 6.81% and 0.972% compared to the rectangular and parallel-type uniform distributions, respectively, while maintaining the same theoretical capacity.

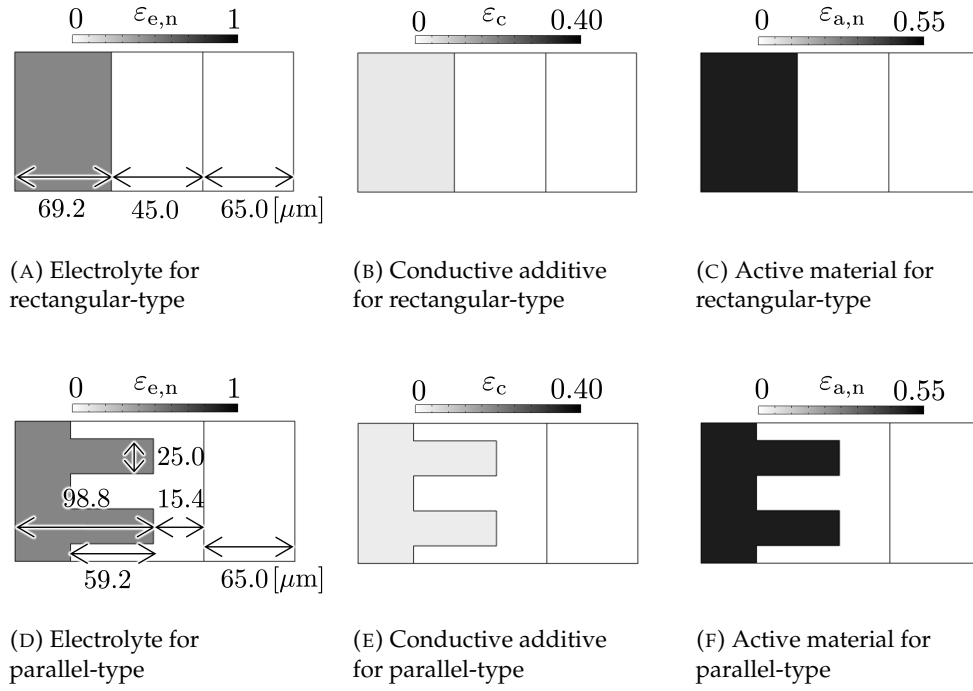


FIGURE 4.18: The uniform material configurations of rectangular-type and parallel-type for the comparison

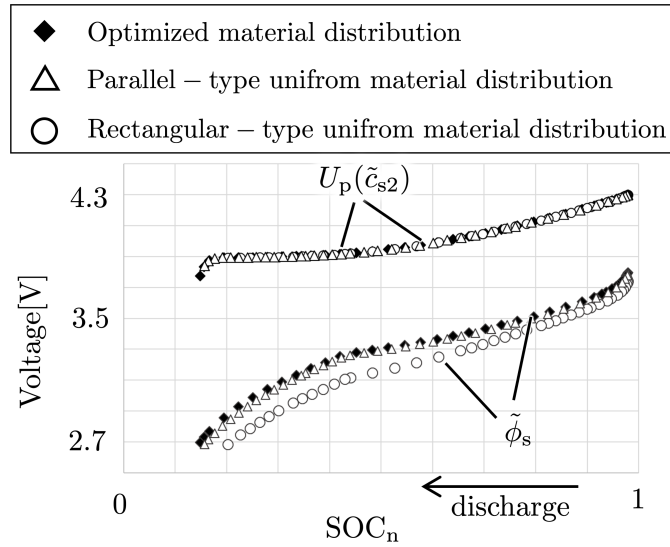


FIGURE 4.19: Histories of $U_p(\tilde{c}_p)$ and discharge curve $\tilde{\phi}_s$ during the discharge process for the uniform rectangular-type, uniform parallel-type and optimized material distributions as functions of SOC_n

Figure 4.19 shows a detailed history of each objective function term during the discharge process. The horizontal axis indicates the SOC of the anode, and the perpendicular axis indicates the voltage. The diamond, triangle, and circle symbols in the graph indicate the history of $U_p(\tilde{c}_p)$ and $\tilde{\phi}_s$ for the optimized configuration, parallel-type uniform material distribution, and rectangular-type uniform material distribution, respectively. As shown in Eq. (4.32), the numerator of the objective function is the difference between $U_p(\tilde{c}_p)$ and $\tilde{\phi}_s$ integrated over time during the discharge process. As the plot shows, the optimized configuration could reduce the difference between the two evaluation variables compared to the uniform distributions.

Here, one can easily criticize that the proposed method exhibits only a slight improvement in the evaluation functions compared to the parallel-type uniform material distribution. However, for a proper comparison from a battery performance perspective, the minimum thickness of the separator must also be considered. The separator is a crucial component in batteries, designed to prevent direct contact between the cathode and anode, thereby avoiding short circuits. From an electrical standpoint, the separator contributes solely to energy loss; thus, minimizing its thickness reduces internal losses within the battery (Li et al., 2024). Based on this consideration, Fig. 4.18d demonstrates that the minimum separator thickness is 15.4 [μm] which is smaller than 45 [μm] in the analysis domain for the optimization. In parallel-type composite anodes, achieving certain electrochemical characteristics requires reducing the separator thickness. In contrast, the proposed method enables the attainment of these characteristics by appropriately distributing the three materials, without the need to reduce the separator thickness. This comparison demonstrates that the proposed method can derive a material distribution that enables battery performance unattainable by the conventional design approach of uniform mixing.

Comparison and analysis of optimal solutions obtained using different material interpolation functions

This paper formulated the interpolation function using microstructure analysis instead of using the SIMP method as the material interpolation function. To demonstrate the importance of the proposed interpolation functions, here we show the comparison between the optimized configuration obtained using the proposed interpolation functions and one obtained using the SIMP method. The material interpolation functions, H_1 and H_2 , can be expressed using the SIMP method as follows:

$$H_1(\varepsilon) = (1 - H_{1,\min})\varepsilon^p + H_{1,\min}, \quad (4.106)$$

$$H_2(\varepsilon_a, \varepsilon_p) = (1 - H_{2,\min})\varepsilon_a^p + K_{\max}(1 - H_{2,\min})\varepsilon_c^p + H_{2,\min}. \quad (4.107)$$

Figure 4.20 illustrates the optimized configurations obtained using the proposed functions and the SIMP method where the penalty parameter p is set to either 3 or 1.5. As can be seen in Fig. 4.20,

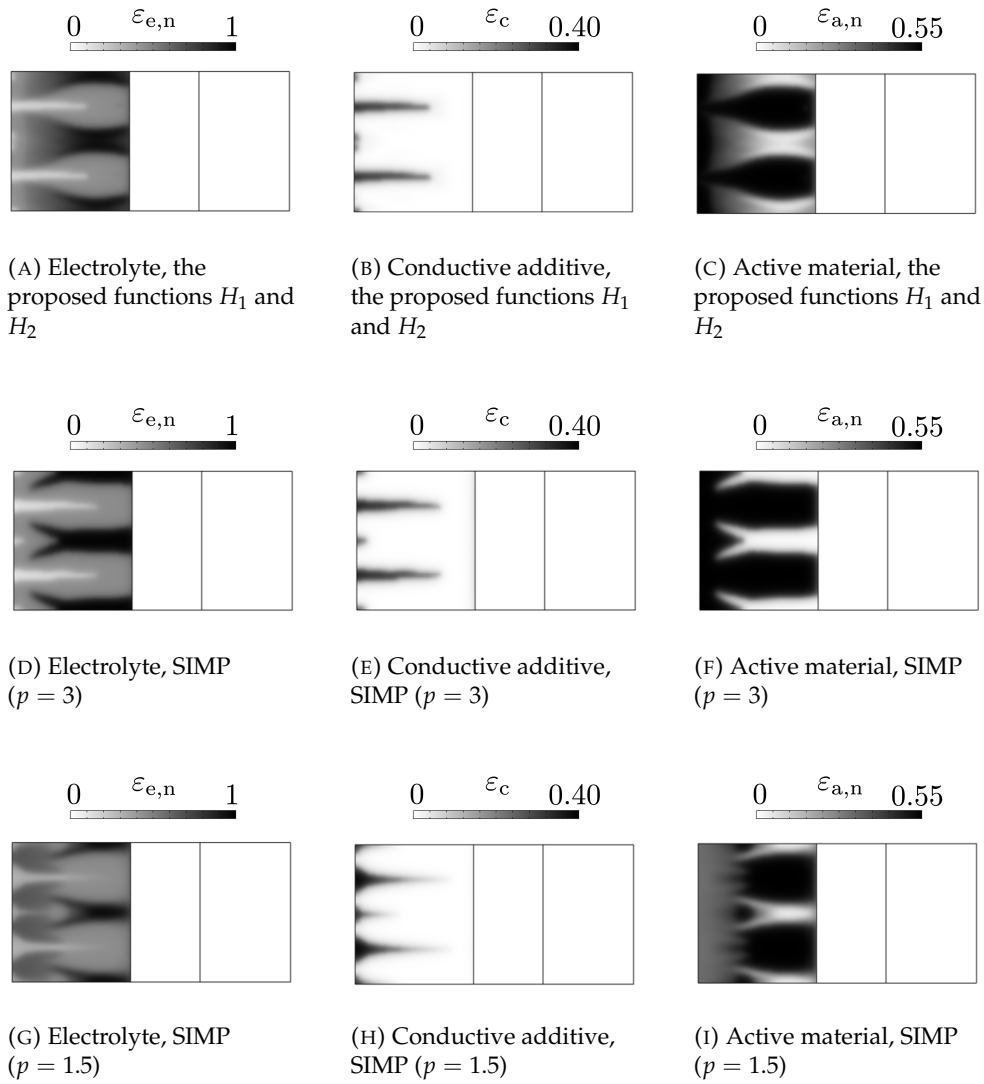


FIGURE 4.20: The obtained material distributions by the optimization method using the proposed function H_1 and H_2 , and the penalty parameter p of 3 and 1.5 with SIMP method

the optimization configuration significantly depends on the interpolation functions used in the optimization calculation. These changes are due to the inclusion of intermediate volume fraction materials in the optimal structure. In other words, in optimization, where design variables are updated based on the gradient of variables, interpolation functions have a significant influence on the direction of the solution search.

Furthermore, the objective function values evaluated using the proposed interpolation function for the optimized configurations obtained by the proposed method and the SIMP method with $p = 3$ and $p = 1.5$ were 2.48, 2.49, and 2.55, respectively. The obtained capacity was 0.831 under all conditions. The objective function value of the optimized configuration obtained using the

proposed method was the smallest, indicating that the proposed optimization method was correctly implemented. The small difference between the optimized configurations obtained using the proposed interpolation function with $R_s = 0.2$ and the SIMP method with $p = 3$ is attributed to the similarity between these interpolation functions, as shown in Fig. 4.12.

Thus, as shown in Fig. 4.20, the optimized configuration strongly depends on the interpolation function profile, and as demonstrated in Fig. 4.12, the interpolation function varies depending on the utilized material diameter. Therefore, when applying the proposed TO method to the design of batteries, it is important to use interpolation functions, H_1 , H_2 and A , that align with their physical interpretations.

Relation between electronic conductivity values of materials and optimized structures

As many researchers devote much effort to materials development, battery performance strongly depends on the physical properties of the materials (Chabot et al., 2013). That is, the physical properties of the materials used in batteries are constantly evolving. For the proposed optimization method to be used in battery development, it is important to show the possibility of deriving the optimized structure with various material properties. Therefore, in this section, the optimized configurations are investigated, setting the various electronic conductivity values of the active material and conductive additive material as (0.001 [S/m], 1 [S/m]), (0.01 [S/m], 10 [S/m]), (0.1 [S/m], 100 [S/m]), and (1 [S/m], 1000 [S/m]). Figure 4.21 shows the obtained optimal material distributions for each pair of electronic conductivities. Comparing Figs. 4.21b, 4.21e, 4.21h and 4.21k, it can be seen that the volume of the conductive additive in the optimized material distribution decreases as the electronic conductivity increases. This tendency is obtained because the resistance due to electron transfer decreases as the electronic conductivity increases, and the volume of the electrolyte and active material can be increased to compensate. Thus, the proposed method can derive the optimal structure for different material properties.

4.6 Conclusions

This paper proposed a TO method that provides the optimized material distribution in the composite anode of ASSBs incorporating microstructure analysis. We achieved the following remarks:

- We formulated an optimization problem to minimize the ratio of the overvoltage, corresponding to the energy loss in the battery charge-discharge reaction, and current. The design variables were set to the normalized densities representing the distributions of the volume fractions of three materials, i.e., electrolyte, conductivity additive, and active materials in the

composite anode, assuming the absence of voids. The design sensitivities used for updating the design variables were derived using the Lagrange and continuous adjoint methods.

- Based on microstructural analysis, material interpolation functions were formulated to describe the relationships between the volume fractions of the materials and their transport coefficients, as well as the relationships between volume fractions and the surface area of the material distributions. To develop these functions, 250 material distributions with a single material type and 420 distributions with two material types were randomly generated with the diameter following the normal distribution on a logarithmic scale. The interpolation functions were then derived using the least-squares method under constraints of monotonicity and positivity. Additionally, the Ersatz material concept was introduced to ensure numerical stability during the optimization process. The validity for determining the order used in the least squares was confirmed using AIC values.
- The optimization algorithm was developed based on the formulation for the optimization problem. The optimized configurations of material distributions in the composite anode represent an unprecedented result that could only be obtained using the TO method. A comparison of the derived optimized distribution with a uniform distribution under the same conditions including the surface area indicated that the proposed method could reduce the overvoltage by 12.4% and enhance the capacity by 6.81%.
- The optimized results obtained using the SIMP method were compared with those from the proposed method to highlight the necessity of appropriate material interpolation functions, which strongly depend on the characteristics of the materials utilized. The obtained distributions were utterly varied, even though the same physical properties were used in the optimization. Namely, it was found that the material interpolation function, which represents the intermediate material's physical properties, significantly affects the optimization when the intermediate material is assumed to be included in the solution.
- The proposed method's effectiveness in battery development, where new materials are developed by various ways and the physical properties of materials evolve daily, was confirmed by changing the electronic conductivity values of the conductive additive material and active material, and deriving the optimized material distributions. As the electronic conductivity increased, the conductive additive's volume in the optimized distributions decreased. This result was obtained because the resistance to electron transfer decreases as conductivity increases, and the volume is replaced by materials that play another role, such as active and electrolyte material. Thus, the proposed method could derive the optimized configurations using a variety of different material properties.

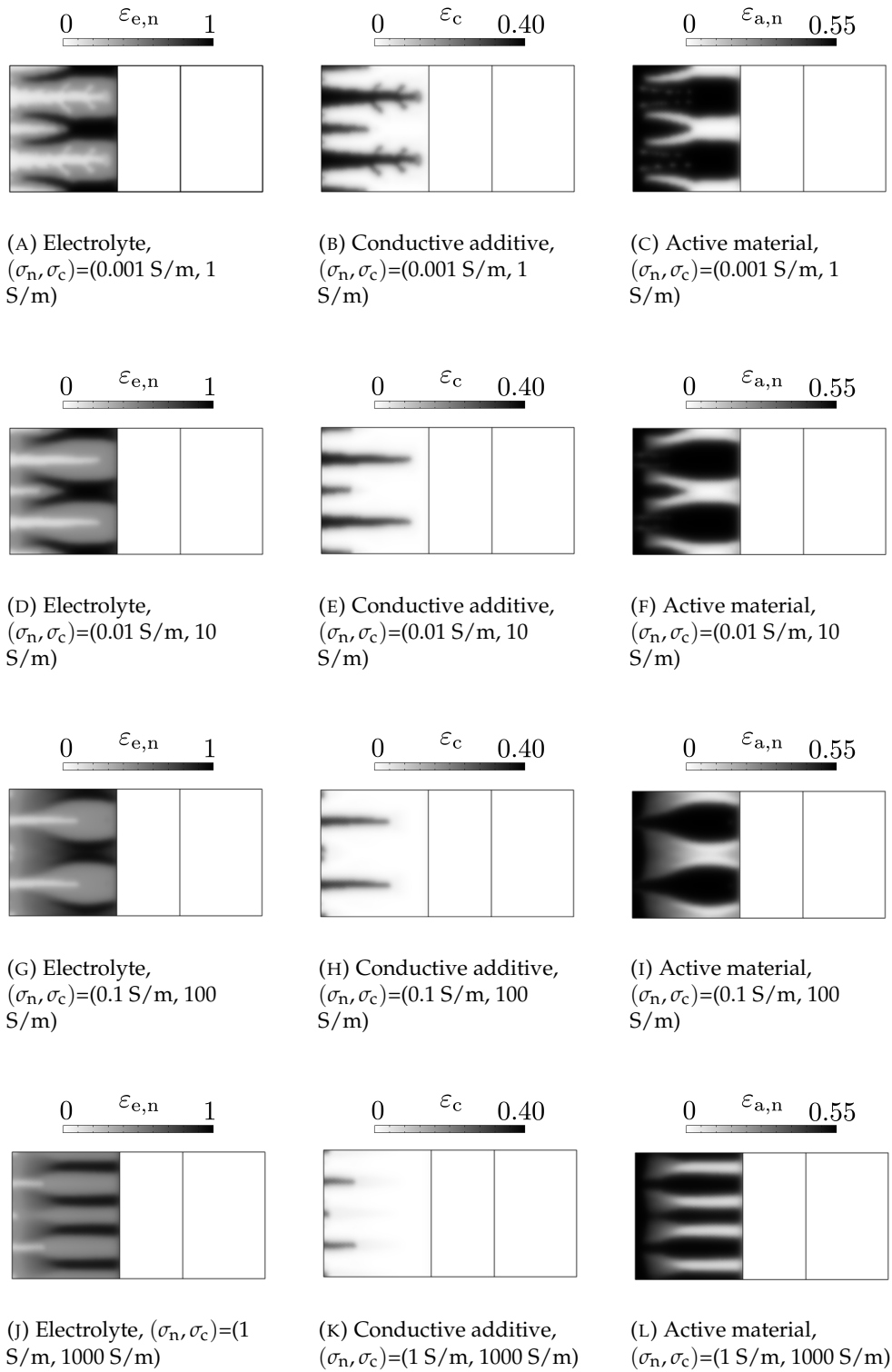


FIGURE 4.21: The obtained material distributions for the various electronic conductivity values of the active material and conductive additive material: $(\sigma_n, \sigma_c) = (0.001 \text{ [S/m]}, 1 \text{ [S/m]})$, $(0.01 \text{ [S/m]}, 10 \text{ [S/m]})$, $(0.1 \text{ [S/m]}, 100 \text{ [S/m]})$, and $(1 \text{ [S/m]}, 1000 \text{ [S/m]})$

Chapter 5

General Conclusions

5.1 Summary of the Dissertation

The escalating global demand for high-performance energy storage, driven by the dual imperatives of decarbonizing the energy sector and electrifying transportation, has created an urgent need for advanced battery technologies. The traditional, empirical-based approach to battery development is increasingly inadequate to meet the diversifying and intensifying performance requirements for applications ranging from grid-scale storage to electric vehicles. In response to this challenge, this dissertation has established a systematic and physics-based design framework for the internal components of secondary batteries, leveraging the principles of topology optimization. The central thesis of this work is that topology optimization, when integrated with sophisticated electrochemical models, can serve as a powerful generative design tool to discover non-intuitive, high-performance battery architectures, thereby accelerating the research and development cycle.

To this end, this research has focused on overcoming two primary obstacles: first, the development of appropriate constitutive laws to link micro- or meso-scale material distributions with macro-scale effective properties governing battery performance; and second, the formulation of computationally efficient sensitivity analysis for time-dependent, coupled multiphysics problems. The methodologies developed were applied to three distinct yet interconnected design problems, progressively increasing in physical complexity and practical relevance. The key contributions and findings of each study are summarized as follows.

In Chapter 2, the research initiated with the design of a lithium-ion battery separator, a critical component for ensuring both safety and performance. The primary objective was to create a separator structure that enhances ionic conductivity while simultaneously suppressing internal short circuits caused by lithium dendrite growth. A novel optimization problem was formulated to maximize the tortuosity of the ion transport path in the direction perpendicular to the electrodes, thereby impeding the linear growth of dendrites. The design sensitivity was derived analytically

using the continuous adjoint method, and its validity was confirmed through numerical examples. The developed optimization algorithm successfully generated two- and three-dimensional separator structures that demonstrated this short-circuit suppression capability. Furthermore, the framework was extended to incorporate practical constraints, demonstrating that by including mechanical stiffness constraints, manufacturable designs connected to fixed boundaries could be obtained, and by including conductivity constraints, optimal solutions could be identified from a pool of high-performance candidates. This chapter established the fundamental applicability of topology optimization to a battery component and laid the groundwork for the more complex, coupled problems addressed subsequently.

In Chapter 3, the scope was extended to the microstructural design of composite electrodes for ASSBs, a promising next-generation technology. This presented a significantly more complex challenge, requiring the consideration of time-dependent, non-linearly coupled electrochemical phenomena. To address this, the homogenization method was employed to link the unit cell's microstructural design to the macro-scale battery performance, governed by the coupled concentration and potential fields. A key innovation of this chapter was the proposal of a novel objective function aimed at achieving a uniform reaction distribution within the composite electrode during discharge. This objective function was shown to be a reliable proxy for standard battery performance metrics while significantly reducing the computational cost of the time-dependent optimization by halving the required time integration domain. The continuous adjoint method was again successfully applied to derive the design sensitivities for this complex, unsteady problem. The resulting optimized microstructures revealed a consistent tendency to form horizontally layered patterns of active material and solid electrolyte, a non-intuitive design that enhances the effective transport properties. This study demonstrated the capability of topology optimization to handle fully coupled, time-dependent battery physics and generate novel microstructural concepts.

In Chapter 4, the research pivoted from microstructural design to a more practical meso-scale optimization of the composite anode in an ASSB, aiming to bridge the gap between theoretical design and manufacturability. Instead of designing the intricate details of a unit cell, the objective became to determine the optimal spatial distribution of the volume fractions of the constituent materials (active material, solid electrolyte, and conductive additive). A central and critical contribution of this chapter was the development of a novel, data-driven methodology for constructing the material interpolation functions that relate local volume fractions to effective transport properties and reaction surface area. This approach, which involved the computational generation and analysis of thousands of random microstructures followed by a constrained least-squares fitting, overcomes the limitations of both the standard SIMP method, which lacks a clear physical basis for intermediate densities, and the homogenization method, which is not applicable when the underlying microstructure is not fixed. The optimization problem was formulated to minimize the overpotential, a direct measure of energy loss. The resulting optimized material distributions, featuring functionally graded compositions, were unprecedented and demonstrated a significant

performance enhancement compared to conventional uniform distributions, reducing overvoltage by 12.4% and increasing usable capacity by 6.81%. This chapter successfully established a new paradigm for the meso-scale design of battery electrodes, providing a practical and powerful tool for engineering next-generation battery architectures.

5.2 Overall Significance and Implications

Collectively, the studies presented in this dissertation make a significant contribution to the field of computational battery design. The work systematically builds a multi-scale topology optimization framework, advancing from a single-physics component design to a fully coupled, time-dependent, multi-material system design. The primary significance of this research lies in its departure from traditional simulation-based analysis towards a new paradigm of physics-based generative design for batteries. Instead of merely predicting the performance of a given design, the developed methodologies autonomously generate novel, high-performance designs from a vast and continuous design space, often leading to non-intuitive solutions that would be difficult to discover through intuition or trial-and-error experimentation.

The implications of this work are twofold. From a scientific perspective, it provides a powerful computational tool to explore the fundamental relationships between a battery's internal structure and its electrochemical performance. It allows for a systematic investigation of how material arrangement at different length scales influences complex phenomena like ion transport, reaction kinetics, and degradation. The data-driven approach for material property modeling proposed in Chapter 4, in particular, offers a flexible and physically grounded way to integrate microstructural information into macro-scale models, a long-standing challenge in the field.

From an engineering and industrial perspective, this research offers a direct pathway to accelerating the development of next-generation batteries. The proposed framework can be used to: (1) rapidly screen and optimize new material combinations, providing clear targets for material scientists; (2) establish design guidelines for advanced manufacturing processes, such as 3D printing or graded electrode fabrication, by providing optimal material volume fraction profiles; and (3) develop application-specific batteries by tailoring the optimization objective and constraints to match specific performance targets (e.g., high power, long life, or high energy density). By reducing the reliance on costly and time-consuming experimental build-and-test cycles, this computational design approach has the potential to significantly shorten the time-to-market for new battery technologies.

5.3 Limitations and Future Research Directions

While this dissertation has successfully established a novel framework for battery design, it is important to acknowledge the assumptions made and to outline directions for future research. The simplifications inherent in the models presented here open up numerous avenues for further investigation to enhance the fidelity and applicability of the proposed methods.

5.3.1 Limitations of the Current Work

The models developed in this thesis were based on several key assumptions:

1. **Isothermal Conditions:** The simulations were performed assuming a constant temperature throughout the battery. In reality, heat generation and dissipation are critical factors affecting performance, degradation, and safety. The coupling between electrochemical and thermal phenomena was not considered.
2. **Neglect of Mechanical Stress:** The models did not account for mechanical stress and strain that arise from volume changes in the active materials during charging and discharging. This is a particularly critical issue in ASSBs, where such stresses can lead to cracking, loss of contact, and failure of conductive pathways.
3. **Absence of Porosity:** In the meso-scale model of Chapter 4, the composite electrode was assumed to be fully dense, with the sum of the material volume fractions equal to one. Real-world electrodes contain pores, which can be either beneficial (e.g., for strain accommodation) or detrimental (e.g., by reducing volumetric energy density). Porosity was not included as a design variable.
4. **Idealized Electrochemistry:** The electrochemical models, while based on established theory, did not include more complex degradation mechanisms such as Solid Electrolyte Interphase (SEI) layer growth, particle cracking, or other side reactions that contribute to capacity fade over a battery's lifetime.

5.3.2 Recommendations for Future Research

Based on these limitations, the following research directions are proposed to build upon the foundation laid by this work:

1. **Fully Coupled Electro-Chemo-Thermal-Mechanical Optimization:** A major and impactful extension of this work would be to develop a fully coupled optimization framework

that incorporates both thermal and mechanical effects. This would involve solving for the temperature and stress fields simultaneously with the electrochemical fields. Such a model would enable the design of structures that not only optimize electrochemical performance but also manage heat generation and mitigate mechanical degradation, leading to safer and longer-lasting batteries.

2. **Optimization Considering Degradation Mechanisms:** Future work should aim to integrate time-dependent degradation models into the optimization framework. By including models for SEI growth or particle fracture, the objective function could be formulated to maximize the battery's cycle life or state of health after a certain number of cycles, rather than just its initial performance. This would represent a significant step towards a true lifetime-aware design methodology.
3. **Multi-Scale Concurrent Optimization:** The current work addressed micro-scale and meso-scale design in separate chapters. A more advanced approach would be to develop a concurrent multi-scale optimization scheme. In such a framework, the meso-scale material distribution (Chapter 4) and the underlying microstructure of the composite material at each point (Chapter 3) would be optimized simultaneously, allowing for a truly hierarchical and holistic design.
4. **Integration of Advanced Manufacturing Constraints:** To further enhance the practical applicability of the generated designs, future research should focus on incorporating more sophisticated manufacturing constraints into the optimization problem. This could include minimum and maximum feature sizes, gradient constraints to ensure smooth transitions in material composition, and constraints that reflect the specific capabilities and limitations of additive manufacturing or other advanced fabrication techniques.
5. **Experimental Validation and Collaboration:** Ultimately, the value of any computational design tool lies in its ability to predict and generate physically realizable and superior products. A crucial future direction is to establish a collaborative loop with experimentalists. The optimized designs generated by the methods in this thesis should be fabricated using advanced manufacturing techniques and their performance should be experimentally characterized. The feedback from this experimental validation would be invaluable for refining the physical models, improving the optimization framework, and ultimately demonstrating the real-world impact of this research.

In conclusion, this dissertation has pioneered the application of topology optimization to the complex, multi-scale, and multiphysics design problems inherent in modern secondary batteries. By establishing a robust and adaptable computational framework, this work provides a new pathway for the systematic design of next-generation energy storage systems, contributing a vital tool in the global effort towards a sustainable energy future.

Appendix A

Sensitivity Analysis

A.1 Minimal heat compliance problem with Helmholtz-type filtering

First, we define the state problem, which describes the physical system. This is a steady-state diffusion equation, where the diffusion coefficient K depends on a filtered density field ρ_H .

$$\begin{aligned} \nabla \cdot \mathbf{q} &= Q && \text{in } \Omega \\ M_q &= \mathbf{q} \cdot \mathbf{n} = q_0 && \text{on } \Gamma_n \\ C &= C_0 && \text{on } \Gamma_d \\ \mathbf{q} &= -K(\rho_H) \nabla C. \end{aligned} \tag{A.1}$$

The material property K is related to a filtered design variable $\bar{\rho}$ through a smoothed Heaviside function, ρ_H . This function ensures a smooth transition of material properties from void (0) to solid (1). The parameter β controls the steepness of this transition.

$$\rho_H = \frac{1}{2} \tanh \left(\beta \left(\bar{\rho} - \frac{1}{2} \right) \right) + \frac{1}{2}. \tag{A.2}$$

A Helmholtz-type filter is applied to the raw design variable ρ to obtain the smoothed (or filtered) design variable $\bar{\rho}$. This regularization is common in topology optimization to ensure mesh-independent solutions and control the minimum length scale of the features. Here, R is the filter radius.

$$\begin{aligned} \nabla \cdot \mathbf{q}_{\bar{\rho}} &= \rho - \bar{\rho} && \text{in } \Omega \\ M_{\bar{\rho}} &= \mathbf{q}_{\bar{\rho}} \cdot \mathbf{n} = q_{\bar{\rho},0} && \text{on } \Gamma_{\rho,n} \\ \bar{\rho} &= \bar{\rho}_0 && \text{on } \Gamma_{\rho,d} \\ \mathbf{q}_{\bar{\rho}} &= -R^2 \nabla \bar{\rho}. \end{aligned} \tag{A.3}$$

The objective functional, f , is defined. This is the quantity we want to minimize or maximize. It is formulated as an integral over the domain and its boundaries, depending on the state variable C .

$$f = \int_{\Omega} QC \, d\Omega - \int_{\Gamma_n} q_0 C \, d\Gamma + \int_{\Gamma_d} M_q C_0 \, d\Gamma. \quad (\text{A.4})$$

To derive the sensitivities, we construct a Lagrangian, $\mathcal{L}$. This is done by augmenting the objective functional f with the weak forms of the state and filter governing equations, multiplied by their respective adjoint variables, $\tilde{C}$ and $\tilde{S}$. The subsequent steps show the application of integration by parts (Green's identity) to shift the differential operators from the state variables $(C, \bar{\rho})$ to the adjoint variables $(\tilde{C}, \tilde{S})$.

$$\mathcal{L} = f + \int_{\Omega} \tilde{C} (\nabla \cdot \mathbf{q} - Q) \, d\Omega + \int_{\Omega} \tilde{S} (\nabla \cdot \mathbf{q}_{\bar{\rho}} - \rho + \bar{\rho}) \, d\Omega \quad (\text{A.5})$$

$$\begin{aligned} &= f + \int_{\Omega} (K \nabla \tilde{C} \cdot \nabla C - \tilde{C} Q) \, d\Omega + \int_{\Gamma} \tilde{C} \mathbf{q} \cdot \mathbf{n} \, d\Gamma \\ &\quad + \int_{\Omega} (R^2 \nabla \tilde{S} \cdot \nabla \bar{\rho} - \tilde{S} \rho + \tilde{S} \bar{\rho}) \, d\Omega + \int_{\Gamma} \tilde{S} \mathbf{q}_{\bar{\rho}} \cdot \mathbf{n} \, d\Gamma \end{aligned} \quad (\text{A.6})$$

$$\begin{aligned} &= f + \int_{\Omega} (C \nabla \cdot \mathbf{q}_a - \tilde{C} Q) \, d\Omega + \int_{\Gamma} \tilde{C} \mathbf{q} \cdot \mathbf{n} \, d\Gamma - \int_{\Gamma} C \mathbf{q}_a \cdot \mathbf{n} \, d\Gamma \\ &\quad + \int_{\Omega} (\bar{\rho} \nabla \cdot \mathbf{q}_{\tilde{S}} - \tilde{S} \rho + \tilde{S} \bar{\rho}) \, d\Omega + \int_{\Gamma} \tilde{S} \mathbf{q}_{\bar{\rho}} \cdot \mathbf{n} \, d\Gamma - \int_{\Gamma} \bar{\rho} \mathbf{q}_{\tilde{S}} \cdot \mathbf{n} \, d\Gamma. \end{aligned} \quad (\text{A.7})$$

We define the adjoint fluxes, $\mathbf{q}_a$ and $\mathbf{q}_{\tilde{S}}$, which are analogous to the state fluxes but are functions of the adjoint variables.

$$\begin{aligned} \mathbf{q}_a &= -K \nabla \tilde{C} \\ \mathbf{q}_{\tilde{S}} &= -R^2 \nabla \tilde{S}. \end{aligned} \quad (\text{A.8})$$

The total variation of the Lagrangian, $\delta \mathcal{L}$, is the sum of its variations with respect to the state variable C , the filtered density $\bar{\rho}$, and the raw density ρ .

$$\delta \mathcal{L} = \delta_{\bar{\rho}} \mathcal{L} + \delta_{\rho} \mathcal{L} + \delta_C \mathcal{L}. \quad (\text{A.9})$$

The variation of $\mathcal{L}$ with respect to the state variable C is computed. Setting $\delta_C \mathcal{L} = 0$ yields the governing equations for the primary adjoint problem.

$$\begin{aligned} \delta_C \mathcal{L} &= \int_{\Omega} (\nabla \cdot \mathbf{q}_a + Q) \delta C \, d\Omega + \int_{\Gamma_d} (\tilde{C} + C_0) \delta M_q \, d\Gamma \\ &\quad - \int_{\Gamma_n} (M_{q,a} + q_0) \delta C \, d\Gamma. \end{aligned} \quad (\text{A.10})$$

Here, $M_{q,a}$ is the normal component of the adjoint flux $\mathbf{q}_a$,

$$M_{q,a} = \mathbf{q}_a \cdot \mathbf{n}. \quad (\text{A.11})$$

Next, the variation of $\mathcal{L}$ with respect to the filtered density $\bar{\rho}$ is derived. This expression contains the sensitivity of the material property K with respect to $\bar{\rho}$. Setting the terms multiplying $\delta\bar{\rho}$ to zero will lead to the second adjoint problem.

$$\begin{aligned} \delta_{\bar{\rho}}\mathcal{L} = & \int_{\Omega} \left(\frac{\partial K}{\partial \rho_H} \frac{\partial \rho_H}{\partial \bar{\rho}} \nabla \tilde{C} \cdot \nabla C \right) \delta\bar{\rho} \, d\Omega - \int_{\Gamma_d} \tilde{C} \frac{\partial K}{\partial \rho_H} \frac{\partial \rho_H}{\partial \bar{\rho}} (\nabla C \cdot \mathbf{n}) \delta\bar{\rho} \, d\Gamma \\ & + \int_{\Omega} (\nabla \cdot \mathbf{q}_{\tilde{S}} + \tilde{S}) \delta\bar{\rho} \, d\Omega + \int_{\Gamma_{\rho,d}} \tilde{S} \delta M_{\bar{\rho}} \, d\Gamma - \int_{\Gamma_{\rho,n}} M_{\tilde{S}} \delta\bar{\rho} \, d\Gamma. \end{aligned} \quad (\text{A.12})$$

Similarly, $M_{\tilde{S}}$ is the normal component of the second adjoint flux $\mathbf{q}_{\tilde{S}}$.

$$M_{\tilde{S}} = \mathbf{q}_{\tilde{S}} \cdot \mathbf{n}. \quad (\text{A.13})$$

Finally, the variation of $\mathcal{L}$ with respect to the raw design variable ρ is found. The term $-\tilde{S}$ represents the sensitivity of the objective functional with respect to the unfiltered design variable.

$$\delta_{\rho}\mathcal{L} = \int_{\Omega} -\tilde{S} \delta\rho \, d\Omega. \quad (\text{A.14})$$

By setting the integrands in $\delta_C\mathcal{L}$ that multiply the variations δC and δM_q to zero, we obtain the strong form of the primary adjoint problem for the adjoint variable $\tilde{C}$. This is a diffusion problem that is solved "backwards" from the objective functional.

$$\begin{aligned} \nabla \cdot \mathbf{q}_a &= -Q && \text{in } \Omega \\ M_{q,a} = \mathbf{q}_a \cdot \mathbf{n} &= -q_0 && \text{on } \Gamma_n \\ \tilde{C} &= -C_0 && \text{on } \Gamma_d \\ \mathbf{q}_a &= -K \nabla \tilde{C}. \end{aligned} \quad (\text{A.15})$$

Similarly, setting the integrands in $\delta_{\bar{\rho}}\mathcal{L}$ that multiply the variations $\delta\bar{\rho}$ and $M_{\bar{\rho}}$ to zero gives the strong form of the second adjoint problem for the adjoint variable $\tilde{S}$. The source terms for this problem depend on the solutions of both the state problem (C) and the first adjoint problem ($\tilde{C}$).

$$\begin{aligned} \nabla \cdot \mathbf{q}_{\tilde{S}} &= -S_{\Omega} - \tilde{S} && \text{in } \Omega \\ M_{\tilde{S}} = \mathbf{q}_{\tilde{S}} \cdot \mathbf{n} &= S_{\Gamma_d} && \text{on } \Gamma_{\rho,n} \cap \Gamma_d \\ M_{\tilde{S}} &= 0 && \text{on } \Gamma_{\rho,n} \cap \Gamma_d^c \\ \tilde{S} &= 0 && \text{on } \Gamma_{\rho,d} \cap \Gamma_d \\ \tilde{S} &= 0 && \text{on } \Gamma_{\rho,d} \cap \Gamma_d^c \\ \mathbf{q}_{\tilde{S}} &= -R^2 \nabla \tilde{S}. \end{aligned} \quad (\text{A.16})$$

The source terms for the second adjoint problem, S_{Ω} and S_{Γ_d} , are defined below. They represent the sensitivity of the state problem's governing equation to changes in the filtered design variable

$\bar{\rho}$.

$$\begin{aligned} S_{\Omega} &= \frac{\partial K}{\partial \rho_H} \frac{\partial \rho_H}{\partial \bar{\rho}} \nabla \tilde{C} \cdot \nabla C \\ S_{\Gamma_d} &= -\tilde{C} \frac{\partial K}{\partial \rho_H} \frac{\partial \rho_H}{\partial \bar{\rho}} (\nabla C \cdot \boldsymbol{n}). \end{aligned} \tag{A.17}$$

A.2 Sensitivity analysis for ASSBs problem

In this appendix, we describe the derivation of the adjoint equation Eq. 3.24. The Lagrangian, defined in Eq. 3.22, is manipulated as follows:

$$\begin{aligned}
\mathcal{L} = & J + \int_{\mathcal{T}} \int_{\Omega_n} \left\{ \frac{\partial (\varepsilon_{s,n} a_1 c_{s,n})}{\partial t} - c_{s,n} \frac{\partial (\varepsilon_{s,n} a_1)}{\partial t} \right. \\
& - \nabla \cdot (a_1 D_{s,n}^{\text{eff}} \nabla c_{s,n}) + \nabla a_1 \cdot (D_{s,n}^{\text{eff}} \nabla c_{s,n}) + \frac{j_n^{\text{Li}}}{F} a_1 \Big\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Omega_p} \left\{ \frac{\partial (\varepsilon_{s,p} a_2 c_{s,p})}{\partial t} - c_{s,p} \frac{\partial (\varepsilon_{s,p} a_2)}{\partial t} \right. \\
& - \nabla \cdot (a_2 D_{s,p}^{\text{eff}} \nabla c_{s,p}) + \nabla a_2 \cdot (D_{s,p}^{\text{eff}} \nabla c_{s,p}) + \frac{j_p^{\text{Li}}}{F} a_2 \Big\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Omega_n} \left\{ \nabla \cdot (a_3 \sigma_{s,n}^{\text{eff}} \nabla \phi_{s,n}) - \nabla a_3 \cdot (\sigma_{s,n}^{\text{eff}} \nabla \phi_{s,n}) - j_n^{\text{Li}} a_3 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Omega_p} \left\{ \nabla \cdot (a_4 \sigma_{s,p}^{\text{eff}} \nabla \phi_{s,p}) - \nabla a_4 \cdot (\sigma_{s,p}^{\text{eff}} \nabla \phi_{s,p}) - j_p^{\text{Li}} a_4 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Omega} \left\{ \nabla \cdot (a_5 \kappa_m^{\text{eff}} \nabla \phi_{e,m}) - \nabla a_5 \cdot (\kappa_m^{\text{eff}} \nabla \phi_{e,m}) + j_m^{\text{Li}} \right\} d\Omega dt \quad (m = n, l, p) \quad (\text{A.18})
\end{aligned}$$

$$\begin{aligned}
= & J + \int_{\mathcal{T}} \int_{\Omega_n} \left\{ \frac{\partial (\varepsilon_{s,n} a_1 c_{s,n})}{\partial t} - c_{s,n} \frac{\partial (\varepsilon_{s,n} a_1)}{\partial t} - \nabla \cdot (a_1 D_{s,n}^{\text{eff}} \nabla c_{s,n}) \right. \\
& + \nabla \cdot (c_{s,n} D_{s,n}^{\text{eff}} \nabla a_1) - c_{s,n} \nabla \cdot (D_{s,n}^{\text{eff}} \nabla a_1) + \frac{j_n^{\text{Li}}}{F} a_1 \Big\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Omega_p} \left\{ \frac{\partial (\varepsilon_{s,p} a_2 c_{s,p})}{\partial t} - c_{s,p} \frac{\partial (\varepsilon_{s,p} a_2)}{\partial t} - \nabla \cdot (a_2 D_{s,p}^{\text{eff}} \nabla c_{s,p}) \right. \\
& + \nabla \cdot (c_{s,p} D_{s,p}^{\text{eff}} \nabla a_2) - c_{s,p} \nabla \cdot (D_{s,p}^{\text{eff}} \nabla a_2) + \frac{j_p^{\text{Li}}}{F} a_2 \Big\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Omega_n} \left\{ \nabla \cdot (a_3 \sigma_{s,n}^{\text{eff}} \nabla \phi_{s,n}) - \nabla \cdot (\phi_{s,n} \sigma_{s,n}^{\text{eff}} \nabla a_3) \right. \\
& + \phi_{s,n} \nabla \cdot (\sigma_{s,n}^{\text{eff}} \nabla a_3) - j_n^{\text{Li}} a_3 \Big\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Omega_p} \left\{ \nabla \cdot (a_4 \sigma_{s,p}^{\text{eff}} \nabla \phi_{s,p}) - \nabla \cdot (\phi_{s,p} \sigma_{s,p}^{\text{eff}} \nabla a_4) \right. \\
& + \phi_{s,p} \nabla \cdot (\sigma_{s,p}^{\text{eff}} \nabla a_4) - j_p^{\text{Li}} a_4 \Big\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Omega} \left\{ \nabla \cdot (a_5 \kappa_m^{\text{eff}} \nabla \phi_{e,m}) - \nabla \cdot (\phi_{e,m} \kappa_m^{\text{eff}} \nabla a_5) \right. \\
& - \phi_{e,m} \nabla \cdot (\kappa_m^{\text{eff}} \nabla a_5) + j_m^{\text{Li}} a_5 \Big\} d\Omega dt \quad (m = n, l, p). \quad (\text{A.19})
\end{aligned}$$

Applying Gauss' theorem, the Lagrangian is further transformed as follows:

$$\begin{aligned}
\mathcal{L} = & J + \int_{\mathcal{T}} \int_{\Omega_n} \left\{ \frac{\partial (\varepsilon_{s,n} a_1 c_{s,n})}{\partial t} - c_{s,n} \frac{\partial (\varepsilon_{s,n} a_1)}{\partial t} - c_{s,n} \nabla \cdot (D_{s,n}^{\text{eff}} \nabla a_1) + \frac{j_n^{\text{Li}}}{F} a_1 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega_n} \left(-a_1 D_{s,n}^{\text{eff}} \nabla c_{s,n} + c_{s,n} D_{s,n}^{\text{eff}} \nabla a_1 \right) \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\Omega_p} \left\{ \frac{\partial (\varepsilon_{s,p} a_2 c_{s,p})}{\partial t} - c_{s,p} \frac{\partial (\varepsilon_{s,p} a_2)}{\partial t} - c_{s,p} \nabla \cdot (D_{s,p}^{\text{eff}} \nabla a_2) + \frac{j_p^{\text{Li}}}{F} a_2 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega_p} \left(-a_2 D_{s,p}^{\text{eff}} \nabla c_{s,p} + c_{s,p} D_{s,p}^{\text{eff}} \nabla a_2 \right) \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\Omega_n} \left\{ \phi_{s,n} \nabla \cdot (\sigma_{s,n}^{\text{eff}} \nabla a_3) - j_n^{\text{Li}} a_3 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega_n} \left(a_3 \sigma_{s,n}^{\text{eff}} \nabla \phi_{s,n} - \phi_{s,n} \sigma_{s,n}^{\text{eff}} \nabla a_3 \right) \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\Omega_p} \left\{ \phi_{s,p} \nabla \cdot (\sigma_{s,p}^{\text{eff}} \nabla a_4) - j_p^{\text{Li}} a_4 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega_p} \left(a_4 \sigma_{s,p}^{\text{eff}} \nabla \phi_{s,p} - \phi_{s,p} \sigma_{s,p}^{\text{eff}} \nabla a_4 \right) \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\Omega} \left\{ \phi_{e,m} \nabla \cdot (\kappa_m^{\text{eff}} \nabla a_5) + j_m^{\text{Li}} \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega} \left(a_5 \kappa_m^{\text{eff}} \nabla \phi_{e,m} - \phi_{e,m} \kappa_m^{\text{eff}} \nabla a_5 \right) \cdot \mathbf{n} d\Gamma dt \\
& (m = n, l, p).
\end{aligned} \tag{A.20}$$

Subsequently, the boundary conditions are applied as:

$$\begin{aligned}
\mathcal{L} = & J + \int_{\Omega_n} [\varepsilon_{s,n} a_1 c_{s,n}]_{t=0}^{t=t_{\max}} d\Omega \\
& + \int_{\mathcal{T}} \int_{\Omega_n} \left\{ -c_{s,n} \frac{\partial (\varepsilon_{s,n} a_1)}{\partial t} - c_{s,n} \nabla \cdot (D_{s,n}^{\text{eff}} \nabla a_1) + \frac{j_n^{\text{Li}}}{F} a_1 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega_n} c_{s,n} D_{s,n}^{\text{eff}} \nabla a_1 \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\Omega_p} \left\{ \frac{\partial (\varepsilon_{s,p} a_2 c_{s,p})}{\partial t} - c_{s,p} \frac{\partial (\varepsilon_{s,p} a_2)}{\partial t} - c_{s,p} \nabla \cdot (D_{s,p}^{\text{eff}} \nabla a_2) + \frac{j_p^{\text{Li}}}{F} a_2 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega_p} c_{s,p} D_{s,p}^{\text{eff}} \nabla a_2 \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\Omega_n} \left\{ \phi_{s,n} \nabla \cdot (\sigma_{s,n}^{\text{eff}} \nabla a_3) - j_n^{\text{Li}} a_3 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Gamma_{\text{left}}} a_3 \sigma_{s,n}^{\text{eff}} \nabla \phi_{s,n} \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega_n \setminus \Gamma_{\text{left}}} (-\phi_{s,n} \sigma_{s,n}^{\text{eff}} \nabla a_3) \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\Omega_p} \left\{ \phi_{s,p} \nabla \cdot (\sigma_{s,p}^{\text{eff}} \nabla a_4) - j_p^{\text{Li}} a_3 \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\Gamma_{\text{right}}} \left(-\frac{I}{A_g} a_4 - \phi_{s,p} \sigma_{s,p}^{\text{eff}} \nabla a_4 \cdot \mathbf{n} \right) d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega_p \setminus \Gamma_{\text{right}}} (-\phi_{s,p} \sigma_{s,p}^{\text{eff}} \nabla a_4) \cdot \mathbf{n} d\Gamma dt \\
& + \int_{\mathcal{T}} \int_{\Omega} \left\{ -\phi_{e,m} \nabla \cdot (\kappa_m^{\text{eff}} \nabla a_5) + j_m^{\text{Li}} \right\} d\Omega dt \\
& + \int_{\mathcal{T}} \int_{\partial\Omega} (-\phi_{e,m} \kappa_m^{\text{eff}} \nabla a_5) \cdot \mathbf{n} d\Gamma dt \quad (m = n, l, p).
\end{aligned} \tag{A.21}$$

The Gâteaux differential with respect to $c_{s,n}$ is taken as follows:

$$\begin{aligned}
d\mathcal{L}(c_{s,n}; \psi) &= \lim_{\xi \rightarrow 0} \frac{\mathcal{L}(c_{s,n} + \xi \psi) - \mathcal{L}(c_{s,n})}{\xi} \\
&= \int_{\Omega_n} \varepsilon_{s,n} a_1|_{t=t_{\max}} d\Omega + \int_{\mathcal{T}} \int_{\Omega_n} -\psi \frac{2}{C_{\text{pr}} t_{\max} V_n} \nabla \cdot (\nabla c_{s,n}) d\Omega dt \\
&\quad + \int_{\mathcal{T}} \int_{\partial\Omega_n} \psi \frac{2}{C_{\text{pr}} t_{\max} V_n} \nabla c_{s,n} \cdot \mathbf{n} d\Gamma dt \\
&\quad + \int_{\Omega_n} \psi \varepsilon_{s,n} a_1(\mathbf{x}, t(t_{\max})) d\Omega \\
&\quad + \int_{\mathcal{T}} \int_{\Omega_n} \psi \left\{ -\frac{\partial (\varepsilon_{s,n} a_1)}{\partial t} - \nabla \cdot (D_{s,n}^{\text{eff}} \nabla a_1) + A \left(-M \frac{\partial U_n}{\partial c_{s,n}} \right) \right\} d\Omega dt \\
&\quad + \int_{\mathcal{T}} \int_{\partial\Omega_n} \psi D_{s,n}^{\text{eff}} \nabla a_1 \cdot \mathbf{n} d\Gamma dt,
\end{aligned} \tag{A.22}$$

where A and M are defined in Eq. (3.25) and Eq. (3.26), respectively. The Gâteaux differentials with respect to $c_{s,p}$, $\phi_{s,n}$, $\phi_{s,p}$, and ϕ_e can be derived as follows:

$$\begin{aligned}
d\mathcal{L}(c_{s,p}; \psi) &= \lim_{\xi \rightarrow 0} \frac{\mathcal{L}(c_{s,p} + \xi\psi) - \mathcal{L}(c_{s,p})}{\xi} \\
&= \int_{\Omega_p} \varepsilon_{s,p} a_2|_{t=t_{\max}} d\Omega + \int_{\Omega_p} \psi \varepsilon_{s,p} a_2(\mathbf{x}, t(t_{\max})) d\Omega \\
&\quad + \int_{\mathcal{T}} \int_{\Omega_p} \psi \left\{ -\frac{\partial(\varepsilon_{s,p} a_2)}{\partial t} - \nabla \cdot (D_{s,p}^{\text{eff}} \nabla a_2) + A \left(-M \frac{\partial U_p}{\partial c_{s,p}} \right) \right\} d\Omega dt \\
&\quad + \int_{\mathcal{T}} \int_{\partial\Omega_p} \psi D_{s,p}^{\text{eff}} \nabla a_2 \cdot \mathbf{n} d\Gamma dt, \tag{A.23}
\end{aligned}$$

$$\begin{aligned}
d\mathcal{L}(\phi_{s,n}; \psi) &= \lim_{\xi \rightarrow 0} \frac{\mathcal{L}(\phi_{s,n} + \xi\psi) - \mathcal{L}(\phi_{s,n})}{\xi} \\
&= \int_{\mathcal{T}} \int_{\Omega_n} \psi \left\{ \nabla \cdot (\sigma_{s,n}^{\text{eff}} \nabla a_3) + AM \right\} d\Omega dt \\
&\quad + \int_{\mathcal{T}} \int_{\Gamma_{\text{left}}} a_3 \sigma_{s,n}^{\text{eff}} \nabla \psi \cdot \mathbf{n} d\Gamma dt \\
&\quad + \int_{\mathcal{T}} \int_{\partial\Omega_n \setminus \Gamma_{\text{left}}} \psi (-\sigma_{s,n}^{\text{eff}} \nabla a_3) \cdot \mathbf{n} d\Gamma dt, \tag{A.24}
\end{aligned}$$

$$\begin{aligned}
d\mathcal{L}(\phi_{s,p}; \psi) &= \lim_{\xi \rightarrow 0} \frac{\mathcal{L}(\phi_{s,p} + \xi\psi) - \mathcal{L}(\phi_{s,p})}{\xi} \\
&= \int_{\mathcal{T}} \int_{\Omega_p} \psi \left\{ \nabla \cdot (\sigma_{s,p}^{\text{eff}} \nabla a_4) + AM \right\} d\Omega dt \\
&\quad + \int_{\mathcal{T}} \int_{\partial\Omega_p} \psi (-\sigma_{s,p}^{\text{eff}} \nabla a_4) \cdot \mathbf{n} d\Gamma dt, \tag{A.25}
\end{aligned}$$

$$\begin{aligned}
d\mathcal{L}(\phi_e; \psi) &= \lim_{\xi \rightarrow 0} \frac{\mathcal{L}(\phi_e + \xi\psi) - \mathcal{L}(\phi_e)}{\xi} \\
&= \int_{\mathcal{T}} \int_{\Omega} \psi \left\{ \nabla \cdot (\kappa_m^{\text{eff}} \nabla a_5) - AM \right\} d\Omega dt \\
&\quad + \int_{\mathcal{T}} \int_{\partial\Omega} \psi (-\kappa_m^{\text{eff}} \nabla a_5) \cdot \mathbf{n} d\Gamma dt. \tag{A.26}
\end{aligned}$$

Finally, Eq. (3.24), Eq. (3.27), Eq. (3.28), Eq. (3.29), and Eq. (3.30), representing the adjoint equation, are derived to satisfy the conditions, $d\mathcal{L}(c_{s,n}; \psi) = d\mathcal{L}(c_{s,p}; \psi) = d\mathcal{L}(\phi_{s,n}; \psi) = d\mathcal{L}(\phi_{s,p}; \psi) = d\mathcal{L}(\phi_e; \psi) = 0$, respectively.

Appendix B

Implementation details

B.1 Random material distribution schemes and estimation of normalized transport coefficient

This appendix presents the finite element analysis code used to determine the diffusion coefficients in microstructural analysis. In this study, the code was implemented using FreeFem++ in conjunction with MPI parallelization. As described in Section 4.3.1, the computational procedure involves randomly generating the logarithm of the radii and the coordinates of the spheres based on a normal distribution, followed by the calculation of fluxes in the x , y , and z directions.

```
// Parallel code for material information
// Naoyuki Ishida
// 2025/01/27
// Conduct code as follows
// mpirun -n 12 FreeFem++-mpi 003_surface_are.edp

// input value [r, ratiomicro]

// 1. Make geometry (output : Temp/micro_mesh.mesh and Temp/rho.sol)
// 2. Solve Heat1~Heat3
// 3. Evaluate D (output : Temp/date_r_ratio_.dat and )

// Caution
// This command affects the results a lot -> srand(10*clock());

load "medit"
load "PETSc" // PETSc plugin
load "iovtk"
load "mshmet"
macro dimension()3// EOM // 2D or 3D
include "macro_ddm.idp" // additional DDM functions
load "ffrandom"
```

```

srandomdev(); //set a true random seed
//warning: under window this command
//change the seed by randinit(random())) so all
//FreeFEM random function are changed

macro def2(i)[i, i#B, i#C]// EOM // vector field definition
macro init2(i)[i, i, i]// EOM // vector field initialization
macro def1(i)i// EOM // scalar field definition
macro init1(i)i// EOM // scalar field initialization
//finite element space
//func Pk = [P1, P1, P1];
func Pk = [P2, P2, P1];
func Pk1= [P1];
func Pk2= [P2,P2,P2];
func Pk3= [P2];
func Pk4= [P0];
mpiComm comm;      // For MPI communication

////////////////////////////////////
// Setting Parameter
////////////////////////////////////

real cpu ; cpu = clock() ;
real Qin1, Qin2, Qin3;
real Qout1, Qout2,Qout3;
real Qin1loc, Qin2loc, Qin3loc;
real Qout1loc, Qout2loc,Qout3loc;

int ratiomicro; ratiomicro = 80 ; //mesh number
real ratio ; ratio = 80. ; //mesh number
real r ; r = 0.10; //radius of particle1
real rave ; rave = log(r)/log(10.) ; //radius of particle1
real rdiv ; rdiv = 0.1; // standard deviation of particle1
real rB ; rB = 0.05; //radius of particle2
real rBave ; rBave = log(rB)/log(10.) ; //radius of particle2
real rBdiv ; rBdiv = 0.1; //standard deviation of particle2
real Dmax ; Dmax = 1.0 ; //Diffusion coefficient for material (particle)
real DBmax ; DBmax = 100 ; //conductibility of conductive additive / that of active
material
real targetvol ; targetvol = 0.00; //target volume of particle //0.01
real targetvolB; targetvolB = 0.00; //target volume of particle //0.01

int pointnum ; pointnum = 10;
int multinum ; multinum = 2;
int maxloop ; maxloop = pointnum * multinum+1;
real multimumreal; multimumreal=multinum;

```

```

real Dmin ; Dmin = Dmax*1e-4;
real cmax ; cmax = 1.0;
real cmin ; cmin = 0.0;
real deltac; deltac = cmin - cmax;
real exD1, exD2, exD3;
real maxrang = 2^31 - 1; //for random parameter
real R ; R = 1/ratio/3.5; //filter radius = 3.5R
real ireal ; ireal = 0;
real lreal ; lreal = 0;

int n = 1.2*targetvol/(4/3*pi*r^3); //number of particle = 1.5 * analytical solution
int M = 1.2*targetvolB/(4/3*pi*rB^3);

real[int,int] position(n,5);
real[int, int] BBB(M, 4);
srandom(10);
//define position

func real normalRandom(real mu, real stddev) {
    real u1 = random()/maxrang, u2 = random()/maxrang;
    real z0 = sqrt(-2.0 * log(u1)) * cos(2.0 * pi * u2); // Box-Muller method
    return mu + z0 * stddev;
}

if(mpirank==0){
    for (int i=0; i<n; i++){
        position(i,0) = random()/maxrang;
        position(i,1) = random()/maxrang;
        position(i,2) = random()/maxrang;
        position(i,4) = normalRandom(rave, rdiv);
        position(i,3) = 10^(position(i,4));
        cout << "- - - - -" << endl;
        cout << " radius = " << position(i,3) << endl;
    }
    // for (int i = 0; i < n; ++i) {
    // cout << "position[" << i << "]" = " << position(i,0)<< ", " << position(i,1)<< ", "
    // << position(i,2) << endl;
    // }

    int count = 0;
    while (count < M) {
        // Generate random coordinate
        real testx = random() / maxrang;
        real testy = random() / maxrang;
        real testz = random() / maxrang;

```

```

real testrBlog = normalRandom(rBave, rBdiv);
real testrB = 10^(testrBlog);

bool isInBeta = true;

// Within Omega or not
for (int i = 0; i < n; i++) {
    real testdx = testx - position(i, 0);
    real testdy = testy - position(i, 1);
    real testdz = testz - position(i, 2);
    real distance = sqrt(testdx*testdx + testdy*testdy + testdz*testdz);

    if (distance <= position(i, 3)) {
        isInBeta = false;
        break;
    }
}

if (isInBeta) {
    BBB(count, 0) = testx;
    BBB(count, 1) = testy;
    BBB(count, 2) = testz;
    BBB(count, 3) = testrB;
    count++;
}
}

broadcast(processor(0),position);
broadcast(processor(0),BBB);

//Create Mesh
include "cube.idp"
// for micro
mesh3 Sh1;          //Mesh of the cubic 1
int [int] NN = [ratiomicro,ratiomicro,ratiomicro]; //Number of mesh for each direction
real [int, int] BB = [[0,1],[0,1],[0,1]]; //the bounding box
int [int, int] L = [[1,7],[3,4],[5,6]]; //Label //Caution!! Level 2 is occupied by the
partition inner boundary
// int[int] r5=[0,2,1,3,7,3,9,3,21,0,13,1];
// Sh1= change(Sh1,region=r5);
if(mpirank==0){
    Sh1 = Cube(NN, BB, L);
}

meshN Sh = Sh1; // Global mesh

```

```

meshN ShNo;          // Non-overlap Mesh of the cubic 1

//copy the Sh from processor "0" to all processor
broadcast(processor(0), Sh);

//partitioning for Sh (Entire Mesh)
macro ShN20()n2o//
int[int] ShN20;
buildDmesh(Sh)
Mat B;  ///!!important
real[int] MC;
{
    createMat(Sh, B, Pk1)    //For Heat
    createPartition(Sh, MC, Pk4)
    fespace Ph1(Sh, Pk4);
    Ph1 part1;
    part1[] = MC;
    ShNo = trunc(Sh, part1 > 0.1);
}

// Make label
int[int] right(1) ; right = [7] ;
int[int] left(1) ; left = [1] ;
int[int] oku(1) ; oku = [4] ;
int[int] temae(1) ; temae = [3] ;
int[int] top(1) ; top = [6] ;
int[int] bottom(1) ; bottom = [5] ;

if(mpirank==0){
    savemesh(Sh, "./Temp/micro_mesh.mesh");
}
// plot(Sh, fill = false, wait = false, cmm="micro_mesh");

//Define fespace and variable
//for micro
/*-----
NOTE:
func Pk = [P1, P1, P1]; // finite element space
func Pk1= [P1];        // finite element space
func Pk2= [P2,P2,P2];  // finite element space
func Pk3= [P2];        // finite element space
func Pk4= [P0];        // finite element space
-----*/

fespace Vh (Sh,Pk1);
fespace Vh2 (Sh,Pk);
Vh<real> def1(D);

```

```

Vh<real> def1(rho), def1(rhoori);
Vh<real> def1(rhoA), def1(rhooriA);
Vh<real> def1(rhoB), def1(rhooriB), def1(rhoBvol);
Vh<real> def1(rhofil), def1(trhofil), def1(rhoHev), def1(rhoHev1);
Vh<real> def1(c1), def1(testc1);
Vh<real> def1(c2), def1(testc2);
Vh<real> def1(c3), def1(testc3);

//Initial guess for rho (active material)
rhooriA = 0 ;
for (int i = 0; i < n; ++i) {
    rhooriA = rhooriA + 1*((x-position(i,0))^2+(y-position(i,1))^2+(z-position(i,2))^2<=(
        position(i,3))^2);
}
rhoA = 1*(rhooriA>=1)+rhooriA*((rhooriA<1)*(rhooriA>0))+0*(rhooriA<=0);

//Initial guess for rhoB (conductive additive material)
rhooriB = 0 ;
for (int i = 0; i < M; ++i) {
    rhooriB = rhooriB + 1*((x-BBB(i,0))^2+(y-BBB(i,1))^2+(z-BBB(i,2))^2<=(BBB(i,3))^2);
}
rhoB = 1*(rhooriB>=1)+rhooriB*((rhooriB<1)*(rhooriB>0))+0*(rhooriB<=0);

rhoori = 0;
rhoori = 1*((rhoB>=1)*(rhoA<0.5)) + rhoB*((rhoB<1)*(rhoB>0.5)*(rhoA<0.5));

rho = 0;
rho = 2 * (rhoori>0.5) + rhoA ;
rho = rho/2;

rhoBvol = 0;
rhoBvol = 1 * (rhoori>0.5) + 0 ;

// Evaluate volume
real volA, volAloc;          // initial normalized volume
volAloc = intN(ShNo)(rhoA);
mpiAllReduce(volAloc, volA, mpiCommWorld, mpiSUM);

real volB, volBloc;          // initial normalized volume
volBloc = intN(ShNo)(rhoB);
mpiAllReduce(volBloc, volB, mpiCommWorld, mpiSUM);

real volt, voltloc;          // initial normalized volume
voltloc = intN(ShNo)(rhoBvol);

```



```

mpiAllReduce(voltloc, volt, mpiCommWorld, mpiSUM);

real beta = 15;
real alpha = 0.5;
//Material interpolate
D = (DBmax - Dmax) * (tanh(beta*alpha)+tanh(beta*((rho-0.5)/(1.0-0.5)) -beta*alpha))/(
    tanh(beta*alpha)+tanh(beta -beta*alpha)) + (Dmax-Dmin)* (tanh(beta*alpha)+tanh(beta
    *((rho-1e-6)/(0.5-1e-6)) -beta*alpha))/(tanh(beta*alpha)+tanh(beta -beta*alpha)) +
    Dmin; // DBmax (rho=1), D_max (rho=0.5), D_min (rho =0)

//Define macros
macro grad(u) [dx(u), dy(u), dz(u)] //
macro nright [1,0,0] //
macro nleft [-1,0,0]//
macro noku [0,1,0] //
macro ntemae [0,-1,0]//
macro ntop [0,0,1] //
macro nbottom[0,0,-1]//

//Define problem
//for micro
varf Filter(def1(rhofil), def1(trhofil)) = intN(Sh,qforder=3)(R^2*grad(rhofil)*grad(
    trhofil)+trhofil*rhofil)
-intN(Sh,qforder=3)(-trhofil*rho)
;

varf Heat1(def1(c1), def1(testc1)) = intN(Sh,qforder=3)(D*grad(testc1)*grad(c1))
+on(right, c1=cmin)
+on(left, c1=cmax)
;
varf Heat2(def1(c2), def1(testc2)) = intN(Sh,qforder=3)(D*grad(testc2)*grad(c2))
+on(oku, c2=cmin)
+on(temae, c2=cmax)
;
varf Heat3(def1(c3), def1(testc3)) = intN(Sh,qforder=3)(D*grad(testc3)*grad(c3))
+on(top, c3=cmin)
+on(bottom, c3=cmax)
;

//Save data
{
    int[int] order = [1,1,1,1,1];
    savevtk("./SaveVTK/000_initial.vtu", ShNo, rhoA, rhoB, rhoori, rho, D, dataname = "
        rhoA rhoB rhoori rho D", order = order);
}

```

```

ireal = 0;
for(int l=0;l<pointnum;l++){

    lreal = l;
    targetvolB = (0.45)*((lreal)/pointnum)+0.0; //Max 0.40
    for(int i=0;i<maxloop;i++){
        if(fmod(i,multimumreal)==1){
            targetvol = (0.60)*(ireal/pointnum);
            ireal =ireal+1;
        }
        if(mpirank==0){
            cout << "+++++++"<<endl;
            cout << " Loop = " << i << ", " << l <<endl;
            cout << " targetvol = " << targetvol << ", " << targetvolB <<endl;
            cout << "+++++++"<<endl;
        }
        //////////////////////////////////////
        //1. Make geometry
        //////////////////////////////////////
        // if(i==0&&j==0){
        //     ofstream f("./Temp/data_radius_"+r+".dat");
        //     f << "step" << " " << "number" << " " << "Radius" << " " << "ln_Radius" << " "
        //         << "average_ln_radius" << " " << "sigma" << "\n";
        // }
        // Create random position
        n = 1.2*targetvol/(4/3*pi*r^3); //number of particle = 1.5 * analytical solution
        M = 1.2*targetvolB/(4/3*pi*rB^3);
        real[int,int] position(n,5);
        real[int, int] BBB(M, 4);
        srand(40*clock());
        if(mpirank==0){
            for (int i=0; i<n; i++){
                position(i,0) = random()/maxrang;
                position(i,1) = random()/maxrang;
                position(i,2) = random()/maxrang;
                position(i,4) = normalRandom(rave, rdiv);
                position(i,3) = 10^(position(i,4));
                // cout << "- - - - -" <<endl;
                // cout << " radius = " << position(i,3) <<endl;
            }
            // for (int i = 0; i < n; ++i) {
            //     cout << "position[" << i << "] = " << position(i,0)<< ", " << position(i,1)
            //         << ", " << position(i,2) << endl;
            // }

            int count = 0;

```

```

while (count < M) {
    // generate random coordinates
    real testx = random() / maxrang;
    real testy = random() / maxrang;
    real testz = random() / maxrang;
    real testrBlog = normalRandom(rBave, rBdiv);
    real testrB = 10^(testrBlog);

    bool isInBeta = true;

    // Within Omega or not
    for (int i = 0; i < n; i++) {
        real testdx = testx - position(i, 0);
        real testdy = testy - position(i, 1);
        real testdz = testz - position(i, 2);
        real distance = sqrt(testdx*testdx + testdy*testdy + testdz*testdz);

        if (distance <= position(i, 3)) {
            isInBeta = false;
            break;
        }
    }

    if (isInBeta) {
        BBB(count, 0) = testx;
        BBB(count, 1) = testy;
        BBB(count, 2) = testz;
        BBB(count, 3) = testrB;
        count++;
    }
}

broadcast(processor(0), position);
broadcast(processor(0), BBB);

// Make rho
rhooriA = 0 ;
for (int i = 0; i < n; ++i) {
    rhooriA = rhooriA + 1*((x-position(i,0))^2+(y-position(i,1))^2+(z-position(i,2))^2<=(position(i,3))^2);
}
rhoA = 1*(rhooriA>=1)+rhooriA*((rhooriA<1)*(rhooriA>0))+0*(rhooriA<=0);

//Initial guess for rhoB (conductive additive material)
rhooriB = 0 ;
for (int i = 0; i < M; ++i) {

```

```

    rhooriB = rhooriB + 1*((x-BBB(i,0))^2+(y-BBB(i,1))^2+(z-BBB(i,2))^2<=(BBB(i,3))^2)
    ;
}
rhoB = 1*(rhooriB>=1)+rhooriB*((rhooriB<1)*(rhooriB>0))+0*(rhooriB<=0);

rhoori = 0;
rhoori = 1*((rhoB>=1)*(rhoA<0.5)) + rhoB*((rhoB<1)*(rhoB>=0.5)*(rhoA<0.5));

rho = 0;
rho = 2 * (rhoori>0.5) + rhoA ;
rho = rho/2;

rhoBvol = 0;
rhoBvol = 1 * (rhoori>0.5) + 0 ;

real volt, voltloc;          // initial normalized volume
voltloc = intN(ShNo)(rhoBvol);
mpiAllReduce(voltloc, volt, mpiCommWorld, mpiSUM);

if(mpirank==0){
    cout << "===== "<<endl;
    cout << " Solve Filter and culculate the surface "<<endl;
    cout << "===== "<<endl;
}

////////////////////////////////////
// For Filtered
////////////////////////////////////
// define matrix & right hand side
real[int] Filterrhs = Filter(0, Vh, tgv=-1.); //right hand side, set "tgv=-1"
B = Filter(Vh, Vh, tgv=-1);
// set(K1, sparams = "-ksp_monitor -pc_type hypre -ksp_max_it 1000 -ksp_type gmres")
;
set(B, sparams = "-pc_type lu"); //Diffusion equation should be "-pc_type lu"
rhofil[] = 0.0;
rhofil[] = B^-1 * Filterrhs;

real surfaceloc, surface;
surfaceloc = intN(ShNo)(sqrt((dx(rhofil))^2+(dy(rhofil))^2+(dz(rhofil))^2)); // Qin
    = -n*(-D*grad) //intN1: surface integral //nright'*grad(c1)
mpiAllReduce(surfaceloc, surface, mpiCommWorld, mpiSUM);

if(mpirank==0){
    cout << "sufrace = " << surface << endl;
}

```

```

}

// rhoHev1 = 30*(rhofil-0.5);
// rhoHev = 1/2.*tanh(rhoHev1)+0.5;
D = (DBmax - Dmax) * (tanh(beta*alpha)+tanh(beta*((rhofil-0.5)/(1.0-0.5)) -beta*
alpha))/(tanh(beta*alpha)+tanh(beta -beta*alpha)) + (Dmax-Dmin)* (tanh(beta*
alpha)+tanh(beta*((rhofil-1e-6)/(0.5-1e-6)) -beta*alpha))/(tanh(beta*alpha)+tanh
(beta -beta*alpha)) + Dmin; // DBmax (rho=1), D_max (rho=0.5), D_min (rho =0)

// Evaluate volume
real volA, volAloc;          // initial normalized volume
volAloc = intN(ShNo)(rhoA);
mpiAllReduce(volAloc, volA, mpiCommWorld, mpiSUM);

real volB, volBloc;          // initial normalized volume
volBloc = intN(ShNo)(rhoB);
mpiAllReduce(volBloc, volB, mpiCommWorld, mpiSUM);

if(mpirank==0){
  cout << "===== "<<endl;
  cout << " Solve Equations " <<endl;
  cout << "===== "<<endl;
}

////////////////////////////////////
// For Heat 1
////////////////////////////////////
// define matrix & right hand side
real[int] Heat1rhs = Heat1(0, Vh, tgv=-1.); //right hand side, set "tgv=-1"
B = Heat1(Vh, Vh, tgv=-1);
// set(K1, sparams = "-ksp_monitor -pc_type hypre -ksp_max_it 1000 -ksp_type gmres")
;
set(B, sparams = "-pc_type lu"); //Diffusion equation should be "-pc_type lu"
c1[] = 0.0;
c1[] = B^-1 * Heat1rhs;

////////////////////////////////////
// For Heat 2
////////////////////////////////////
// define matrix & right hand side
real[int] Heat2rhs = Heat2(0, Vh, tgv=-1.); //right hand side, set "tgv=-1"
B = Heat2(Vh, Vh, tgv=-1);
// set(K1, sparams = "-ksp_monitor -pc_type hypre -ksp_max_it 1000 -ksp_type gmres")
;
set(B, sparams = "-pc_type lu"); //Diffusion equation should be "-pc_type lu"

```

```

c2[] = 0.0;
c2[] = B^-1 * Heat2rhs;

////////////////////////////////////
// For Heat 3
////////////////////////////////////
// define matrix & right hand side
real[int] Heat3rhs = Heat3(0, Vh, tgv=-1.); //right hand side, set "tgv=-1"
B = Heat3(Vh, Vh, tgv=-1);
// set(K1, sparams = "-ksp_monitor -pc_type hypre -ksp_max_it 1000 -ksp_type gmres")
;
set(B, sparams = "-pc_type lu"); //Diffusion equation should be "-pc_type lu"
c3[] = 0.0;
c3[] = B^-1 * Heat3rhs;

Qout1loc = intN1(ShNo,right)(-1*D*nright'*grad(c1));
mpiAllReduce(Qout1loc, Qout1, mpiCommWorld, mpiSUM);

Qout2loc = intN1(ShNo,oku)(-1*D*noku'*grad(c2));
mpiAllReduce(Qout2loc, Qout2, mpiCommWorld, mpiSUM);
Qout3loc = intN1(ShNo,top)(-1*D*ntop'*grad(c3));
mpiAllReduce(Qout3loc, Qout3, mpiCommWorld, mpiSUM);

// Evaluate volume
real Q1, Q1loc; // Q_1
Q1loc = intN(ShNo)(D*grad(c1)'*grad(c1));
mpiAllReduce(Q1loc, Q1, mpiCommWorld, mpiSUM);
// Evaluate volume
real Q2, Q2loc; // Q_1
Q2loc = intN(ShNo)(D*grad(c2)'*grad(c2));
mpiAllReduce(Q2loc, Q2, mpiCommWorld, mpiSUM);
// Evaluate volume
real Q3, Q3loc; // Q_1
Q3loc = intN(ShNo)(D*grad(c3)'*grad(c3));
mpiAllReduce(Q3loc, Q3, mpiCommWorld, mpiSUM);

Qin1loc = intN1(ShNo,left)(D*nleft'*grad(c1)); // Qout = n*(-D*grad)
mpiAllReduce(Qin1loc, Qin1, mpiCommWorld, mpiSUM);
Qin2loc = intN1(ShNo,temae)(D*ntemae'*grad(c2)); // Qout = n*(-D*grad)
mpiAllReduce(Qin2loc, Qin2, mpiCommWorld, mpiSUM);
Qin3loc = intN1(ShNo,bottom)(D*nbottom'*grad(c3)); // Qout = n*(-D*grad)
mpiAllReduce(Qin3loc, Qin3, mpiCommWorld, mpiSUM);

if(mpirank==0){
    // cout << "Qin1 = " << Qin1 << ", Qout1 = " << Qout1 << endl;

```

```

// cout << "Qout1 top = " << Qout11 << ", Qout1 bottom = " << Qout12 << endl;
// cout << "Qout1 oku = " << Qout13 << ", Qout1 temae = " << Qout14 << endl;
cout << "Qin1 = " << Qin1 << ", Qout1 = " << Qout1 << ", Q1 = " << Q1 << endl;
cout << "Qin2 = " << Qin2 << ", Qout2 = " << Qout2 << ", Q2 = " << Q2 << endl;
cout << "Qin3 = " << Qin3 << ", Qout3 = " << Qout3 << ", Q3 = " << Q3 << endl;
exD1 = Qin1/(-1*deltac);
exD2 = Qin2/(-1*deltac);
exD3 = Qin3/(-1*deltac);
cout << "material D = " << Dmax << ", Expected D_x = " << exD1<< ", Expected D_y =
      " << exD2 << ", Expected D_z = " << exD3 << endl;
cout << "Target_Vol_rhoA = " << targetvol << ", Actual_Vol_rhoA = " << volA << "
      Target_Vol_rhoB = " << targetvolB << ", Actual_Vol_rhoB = " << volt << endl;
}
if(mpirank==0){
    cout << "===== " << endl;
    cout << "CPU time = " << (clock()-cpu) << endl;
    cout << "===== " << endl;
}

//Save data
if(fmod(i,5)==1){
    int[int] order = [1,1,1,1,1,1];
    savevtk("./SaveVTK/vtudata_r_"+r+"_meshratio_"+ratiomicro+"_iteri_"+i+"_iterl_"+l+
        "_DBmax_"+DBmax+".vtu", ShNo, rho, D, rhoA, rhoB, rhoori, rhofil, dataname = "
        rho D rhoA rhoB rho_ori filtered_rho", order = order, append = i > 0 ? true :
        false);
}
if(mpirank==0&&i==0&&l==0){
    ofstream f("./Temp/data_r_"+r+"_meshratio_"+ratiomicro+"_DBmax_"+DBmax+".dat",
        append);
    f << "step" << " " << "Mesh_number" << " " << "nondimensionalized_r" << " " << "
        nondimensionalized_rB" << " " << "target_volA" << " " << "actual_volA" << " "
        << "target_volB" << " " << "actual_volB" << " " << "D_max" << " " << "D_max_B"
        << " " << "expected_D1" << " " << "expected_D2" << " " << "expected_D3"<< " " << "
        surface_Area" << " " << "expected_Q1" << " " << "expected_Q2" << " " << "
        expected_Q3"<< " " << "Total_volumeB"<< "\n";
}
if(mpirank==0){
    ofstream f("./Temp/data_r_"+r+"_meshratio_"+ratiomicro+"_DBmax_"+DBmax+".dat",
        append);
    f << i << " " << ratiomicro << " " << r << " " << rB << " " << targetvol << " " <<
        volA << " " << targetvolB << " " << volB << " " << Dmax << " " << DBmax << " "
        << exD1 << " " << exD2 << " " << exD3 << " " << surface << " " << Q1 << " " << Q2
        << " " << Q3 << " "<< volt << "\n";
}
}
}

```

```

    ireal = 0;
}

```

B.2 Least square with constraint

This section presents the code used to construct a constrained interpolation function based on the dataset generated in Appendix B.1. MATLAB was utilized for this implementation. By executing the following code using the ".data" files exported in Appendix B.1 as input arguments, the optimal degree and coefficients for the polynomial approximation are determined based on the Akaike Information Criterion (AIC). It should be noted that the function codes provided subsequently must be saved within a folder named "function" located in the same directory as the main execution script.

First, the code for the single-argument case is presented.

```

clear;
addpath("function/");
% Import data from "test.dat"
data = readmatrix('test.dat');
for n=n_min:n_max
    x_data = data(:, 5);
    x_data = [x_data;1];
    y_data = sqrt(sum(data(:, 11:13).^2, 2))/sqrt(3); % Diffusion
        parameter
    y_data = [y_data;1];
    C = createPolynomialMatrix1D(x_data, n);
    % Create coefficient for constraint
    A1 = createPolynomialMatrix1D_dx(x_data, n);
    A = [A1; -C];
    b = zeros(2*length(x_data),1);
    a = lsqlin(C,y_data,A,b);
    % Conduct reast square
    coeff = C \ y_data;
    % coeff=[a00,a01,...,a0m,a10,a21,...,a2m,...,an1,...,anm]

    % scatter plot
    figure;
    scatter(x_data, y_data, 'filled');
    hold on;
    x_fit = linspace(min(x_data), max(x_data), 1000);
    y_fit = evaluatePolynomial1D(a, x_fit, n);
    plot(x_fit, y_fit, 'r-', 'LineWidth', 2);

```



```

y_coeff = evaluatePolynomial1D(coeff, x_fit, n);
plot(x_fit, y_coeff, 'g-', 'LineWidth', 2);
xlabel('x');
ylabel('y');
title('Scatter Plot and Polynomial Fit');
legend('Data', 'LS with constraint', 'Normal LS');
grid on;
hold off;

% Calculate Res
Y_pred = evaluatePolynomial1D(a, x_data, n); % Predicted values from
      model
RSS = sum((y_data - Y_pred).^2); % Res
num_data = length(y_data); % The number of data
num_params = length(a); % The number of parameter in the model

% AIC Calculation
AIC(n) = 2 * num_params + num_data * log(RSS / num_data);

% Modifiedn AIC (Option)
if num_data > num_params + 1
C_AIC(n) = AIC(n) + (2 * num_params * (num_params + 1)) / (num_data -
      num_params - 1);
else
C_AIC(n) = NaN;
warning('The number of data is insufficient');
end

% Display result
fprintf('n = %d \n', n);
fprintf('AIC: %.4f\n', AIC(n));
fprintf('C-AIC: %.4f\n', C_AIC(n));
poly_str = createPolynomialString(a, n);
poly_str2 = createPolynomialString(coeff, n);
disp("With constraint");
disp(poly_str);
disp("Without constraint");
disp(poly_str2);

% Option
% if AIC(n)<AICMin
%     AICMin = AIC(n);
%     AICMinN = n;
%     AICMinpoly_str2 = poly_str2;
% end
end

```

The following function should be defined in "function" folder.

```
function A = createPolynomialMatrix1D(x_data, n)
    % Inpute:
    % x_data:
    % n: the number of maximum order
    % Output:
    % A: Polynomial Fitting Matrix

    num_data = length(x_data);

    % Initialize matrix A
    A = zeros(num_data, (n + 1));
    col = 1;
    for i = 0:n
        A(:, i+1) = (x_data.^i);
    end
end

function A = createPolynomialMatrix1D_dx(x_data, n)
    % Inpute:
    % x_data:
    % n: the number of maximum order
    % Output:
    % A: Polynomial Fitting Matrix

    num_data = length(x_data);

    % Initialize matrix A
    A = zeros(num_data, (n+1));
    A(:,1) = 0;
    for i = 1:n
        A(:, i+1) = - i * (x_data.^(i-1));
    end
end

function Z_fit = evaluatePolynomial1D(a, x_fit, n)
    % Input:
    % coeff:
    % X_fit, Y_fit:
    % n: The number of mamimum orfer
    % Output:
    % Z_fit:

    % Initialize Z_fit
    Z_fit = zeros(size(x_fit));
```

```
% Calculate Z_fit
for i = 0:n
    Z_fit = Z_fit + a(i+1) * x_fit.^i;
end
end
function poly_str = createPolynomialString(coeff, n)
% Initialization
poly_str = "";
coeff_idx = 1;

for i = 0:n
    % Current coefficient
    current_coeff = coeff(i+1);
    if i == 0
        if current_coeff < 0
            poly_str = poly_str + sprintf(" - %g", abs(current_coeff));
        else
            poly_str = poly_str + sprintf("%g", current_coeff);
        end
    else
        if current_coeff < 0
            poly_str = poly_str + sprintf(" - %g*x^%d", abs(current_coeff),
                i);
        else
            poly_str = poly_str + sprintf(" + %g*x^%d", current_coeff, i);
        end
    end
end
end
end
```

Next, the code for the two-argument case is presented.

```
clear;
% Import data from "test.dat"
data = readmatrix('testdat');

% Please modify to mach your code
x_data = data(:, 6);
y_data = data(:, 18);
z_data = sqrt(sum(data(:, 15:17).^2, 2))/sqrt(3); %Diffusion

for n = n_min:n_max      % Order for x
    for m = m_min:m_max  %Order for y
        % Derive the coefficients using least square method with constraint
        C = createPolynomialMatrix(x_data, y_data, n, m);
        A1 = createPolynomialMatrix_dx(x_data, y_data, n, m);
        A2 = createPolynomialMatrix_dy(x_data, y_data, n, m);
        A = [A1;A2];
        F = [A1;A2;-C];
        b = zeros(3*length(x_data),1);
        a = lsqlin(C,z_data,F,b);

        %Derive the coefficients using least square method without
            constraint
        coeff = C \ z_data; % coeff=[a00,a01,...,a0m,a10,a21,...,a2m,...,an1
            ...,anm]

        % Plot
        [X_fit, Y_fit] = meshgrid(linspace(min(x_data), max(x_data), 50),
            linspace(min(y_data), max(y_data), 50));
        Z_fit = evaluatePolynomial(a, X_fit, Y_fit, n, m);
        mask_fit = Y_fit <= -X_fit + 1;
        Z_fit(~mask_fit) = NaN;

        figure(1);
        h = surf(X_fit, Y_fit, Z_fit);
        alpha(h, 0.5); % Opaciticy
        hold on;
        scatter3(x_data, y_data, z_data, 80, 'k', 'filled');
        scatter3(x_data, y_data, z_data, 30, 'w', 'filled');

        xlabel('Active material');
        ylabel('Conductive additive');
        zlabel('Electrical conductivity');
        title('Least square with constraint');
        grid on;
```

```

hold off;

%%% AIC evalustion%%%
Z_pred = evaluatePolynomial(a, x_data, y_data, n, m);
RSS = sum((z_data - Z_pred).^2);

num_data = length(z_data);
num_params = length(a);
AIC(n,m) = 2 * num_params + num_data * log(RSS / num_data);

% Display result
fprintf('n = %d, m = %d \n', n, m);
fprintf('AIC: %.4f\n', AIC(n,m));
poly_str = createPolynomialString(a, n, m);
poly_str2 = createPolynomialString(coeff, n, m);
disp("With constraint");
disp(poly_str);
disp("Without constraint");
disp(poly_str2);
end
end
saveas(gcf, 'plot.png'); % Save as png

```

Function list:

```

function A = createPolynomialMatrix(x_data, y_data, n, m)
% Input:
% x_data, y_data: data
% n: The number of maximum order for x
% m: The number of maximum order for y
% Output:
% A: Polynomial Fitting Matrix

num_data = length(x_data);

% Initialize matrix A
A = zeros(num_data, (n + 1) * (m + 1));

col = 1;
for i = 0:n
    for j = 0:m
        A(:, col) = (x_data.^i) .* (y_data.^j);
        col = col + 1;
    end
end
end
end

```

```

function A = createPolynomialMatrix_dx(x_data, y_data, n, m)
    num_data = length(x_data);

    % Initialize matrix A
    A = zeros(num_data, (n + 1) * (m + 1));

    col = 1;
    for i = 0:n
        for j = 0:m
            if i == 0
                A(:, col) = 0;
            else
                A(:, col) = - (i) * (x_data.^(i-1)) .* (y_data.^j);
            end
            col = col + 1;
        end
    end
end

function A = createPolynomialMatrix_dy(x_data, y_data, n, m)
    num_data = length(x_data);

    % Initialize matrix A
    A = zeros(num_data, (n + 1) * (m + 1));

    col = 1;
    for i = 0:n
        for j = 0:m
            if j == 0
                A(:, col) = 0;
            else
                A(:, col) = - (j) * (x_data.^(i)) .* (y_data.^(j-1));
            end
            col = col + 1;
        end
    end
end

function poly_str = createPolynomialString(coeff, n, m)
    % Initialization
    poly_str = "";
    coeff_idx = 1;

    % x
    for i = 0:n

```

```
% y
y_terms = "";

% y
for j = 0:m
    current_coeff = coeff(coeff_idx);
    if j == 0
        if current_coeff < 0
            y_terms = y_terms + sprintf(" - %g", abs(current_coeff));
        else
            y_terms = y_terms + sprintf("%g", current_coeff);
        end
    else
        if current_coeff < 0
            y_terms = y_terms + sprintf(" - %g*y^%d", abs(current_coeff),
                j);
        else
            y_terms = y_terms + sprintf(" + %g*y^%d", current_coeff, j);
        end
    end
    coeff_idx = coeff_idx + 1;
end

if i == 0
    poly_str = poly_str + y_terms;
else
    if i == 1
        poly_str = poly_str + sprintf(" + x*(%s)", y_terms);
    else
        poly_str = poly_str + sprintf(" + x^%d*(%s)", i, y_terms);
    end
end
end
end
```


References

- Alizadeh, Mehrzad et al. (2023). "A Numerical Simulation of Evolution Processes and Entropy Generation for Optimal Architecture of an Electrochemical Reaction-Diffusion System: Comparison of Two Optimization Strategies". In: *Journal of The Electrochemical Society* 170.11, p. 114520. DOI: [10.1149/1945-7111/ad0a7c](https://doi.org/10.1149/1945-7111/ad0a7c).
- Allaire, Grégoire (1992). "Homogenization and two-scale convergence". In: *SIAM Journal on Mathematical Analysis* 23.6, pp. 1482–1518. DOI: [10.1137/0523084](https://doi.org/10.1137/0523084).
- Allaire, Grégoire and François Jouve (2005). "A level-set method for vibration and multiple loads structural optimization". In: *Computer Methods in Applied Mechanics and Engineering* 194.30-33, pp. 3269–3290. DOI: [10.1016/j.cma.2004.12.018](https://doi.org/10.1016/j.cma.2004.12.018).
- Allaire, Grégoire et al. (1997). "Shape optimization by the homogenization method". In: *Numerische Mathematik* 76, pp. 27–68.
- Allaire, Grégoire et al. (2004). "Structural optimization using sensitivity analysis and a level-set method". In: *Journal of Computational Physics* 194.1, pp. 363–393.
- Antony Jose, Subin et al. (2025). "Solid-State Lithium Batteries: Advances, Challenges, and Future Perspectives". In: *Batteries* 11.3. ISSN: 2313-0105. DOI: [10.3390/batteries11030090](https://doi.org/10.3390/batteries11030090).
- Arunachalam, Harikesh and Simona Onori (2019). "Full homogenized macroscale model and pseudo-2-dimensional model for lithium-ion battery dynamics: comparative analysis, experimental verification and sensitivity analysis". In: *Journal of The Electrochemical Society* 166.8, A1380. DOI: [10.1149/2.0051908jes](https://doi.org/10.1149/2.0051908jes).
- Arunachalam, Harikesh et al. (2015). "On Veracity of Macroscopic Lithium-Ion Battery Models". In: *Journal of The Electrochemical Society* 162.10, A1940. DOI: [10.1149/2.0771509jes](https://doi.org/10.1149/2.0771509jes).
- Bates, Alex M et al. (2022). "Are solid-state batteries safer than lithium-ion batteries?" In: *Joule* 6.4, pp. 742–755.
- Beck, Victor A. et al. (2021). "Computational design of microarchitected porous electrodes for redox flow batteries". In: *Journal of Power Sources* 512, p. 230453. ISSN: 0378-7753. DOI: [10.1016/j.jpowsour.2021.230453](https://doi.org/10.1016/j.jpowsour.2021.230453).
- Bendsøe, Martin P (1989). "Optimal shape design as a material distribution problem". In: *Structural Optimization* 1, pp. 193–202. DOI: [10.1007/BF01650949](https://doi.org/10.1007/BF01650949).
- Bendsøe, Martin P and Ole Sigmund (1999). "Material interpolation schemes in topology optimization". In: *Archive of Applied Mechanics* 69, pp. 635–654. DOI: [10.1007/s004190050248](https://doi.org/10.1007/s004190050248).
- Bendsøe, Martin P. and Ole Sigmund (2004). *Topology optimization: theory, methods, and applications*. Springer Berlin, Germany.

- Bendsøe, Martin Philip and Noboru Kikuchi (1988). "Generating optimal topologies in structural design using a homogenization method". In: *Computer Methods in Applied Mechanics and Engineering* 71.2, pp. 197–224.
- Bendsøe, Martin Philip and Noboru Kikuchi (1988). "Generating optimal topologies in structural design using a homogenization method". In: *Computer Methods in Applied Mechanics and Engineering* 71.2, pp. 197–224. ISSN: 0045-7825. DOI: [10.1016/0045-7825\(88\)90086-2](https://doi.org/10.1016/0045-7825(88)90086-2).
- Borrvall, Thomas and Joakim Petersson (2003). "Topology optimization of fluids in Stokes flow". In: *International journal for numerical methods in fluids* 41.1, pp. 77–107.
- Box, George EP and Mervin E Muller (1958). "A note on the generation of random normal deviates". In: *The annals of mathematical statistics* 29.2, pp. 610–611.
- Cano, Zachary P et al. (2018). "Batteries and fuel cells for emerging electric vehicle markets". In: *Nature Energy* 3.4, pp. 279–289. DOI: [10.1038/s41560-018-0108-1](https://doi.org/10.1038/s41560-018-0108-1).
- Chabot, Victor et al. (2013). "Effect of electrode physical and chemical properties on lithium-ion battery performance". In: *International Journal of Energy Research* 37.14, pp. 1723–1736.
- Chen, Chih-Hsiang et al. (2019). "Computational design of flow fields for vanadium redox flow batteries via topology optimization". In: *Journal of Energy Storage* 26, p. 100990. DOI: [10.1016/j.est.2019.100990](https://doi.org/10.1016/j.est.2019.100990).
- Chen, Fan et al. (2022). "Topology optimization design and numerical analysis on cold plates for lithium-ion battery thermal management". In: *International Journal of Heat and Mass Transfer* 183, p. 122087. ISSN: 0017-9310. DOI: [10.1016/j.ijheatmasstransfer.2021.122087](https://doi.org/10.1016/j.ijheatmasstransfer.2021.122087).
- Chen, Zhou and Cheng Zhang (2024). "Engineering electrode microstructures for advanced lithium-ion batteries". In: *Microstructures* 4.3. ISSN: 2770-2995. DOI: [10.20517/microstructures.2023.89](https://doi.org/10.20517/microstructures.2023.89).
- Choi, Jae Seok et al. (2011). "Topology optimization using a reaction–diffusion equation". In: *Computer Methods in Applied Mechanics and Engineering* 200.29, pp. 2407–2420. ISSN: 0045-7825. DOI: [10.1016/j.cma.2011.04.013](https://doi.org/10.1016/j.cma.2011.04.013).
- Choi, Junghyun and Patrick Joohyun Kim (2022). "A roadmap of battery separator development: Past and future". In: *Current Opinion in Electrochemistry* 31, p. 100858.
- Chojkiewicz, Emilia and Amol Phadke (2024). "Economic Case for Replacing High-Emitting Peaker Plants with Fuel Cells for Automotive Applications". In: *ACS Omega* 9.34, pp. 36445–36452. DOI: [10.1021/acsomega.4c04227](https://doi.org/10.1021/acsomega.4c04227).
- Chung, Ding-Wen et al. (2013). "Validity of the Bruggeman relation for porous electrodes". In: *Modelling and Simulation in Materials Science and Engineering* 21.7, p. 074009. DOI: [10.1088/0965-0393/21/7/074009](https://doi.org/10.1088/0965-0393/21/7/074009).
- Clausnitzer, Moritz et al. (2023). "Optimizing the Composite Cathode Microstructure in All-Solid-State Batteries by Structure-Resolved Simulations". In: *Batteries & Supercaps* 6.11, e202300167. DOI: [10.1002/batt.202300167](https://doi.org/10.1002/batt.202300167).
- Danilov, D. et al. (2010). "Modeling All-Solid-State Li-Ion Batteries". In: *Journal of The Electrochemical Society* 158.3, A215. DOI: [10.1149/1.3521414](https://doi.org/10.1149/1.3521414).

- Dapogny, C. et al. (2014). "Three-dimensional adaptive domain remeshing, implicit domain meshing, and applications to free and moving boundary problems". In: *Journal of Computational Physics* 262, pp. 358–378. ISSN: 0021-9991. DOI: [10.1016/j.jcp.2014.01.005](https://doi.org/10.1016/j.jcp.2014.01.005).
- Deng, Jiaojiao et al. (2024). "Functionally gradient materials for sustainable and high-energy rechargeable lithium batteries: Design principles, progress, and perspectives". In: *Journal of Energy Chemistry* 99, pp. 426–449. ISSN: 2095-4956. DOI: [10.1016/j.jechem.2024.07.062](https://doi.org/10.1016/j.jechem.2024.07.062).
- Deysher, Grayson et al. (2022). "Transport and mechanical aspects of all-solid-state lithium batteries". In: *Materials Today Physics*.
- Doyle, Marc et al. (1993). "Modeling of galvanostatic charge and discharge of the lithium/polymer/insertion cell". In: *Journal of the Electrochemical Society* 140.6, p. 1526.
- Ebner, Martin et al. (2014). "Tortuosity Anisotropy in Lithium-Ion Battery Electrodes." In: *Advanced Energy Materials* 4.5.
- Feppon, Florian (2024). "Multiscale Topology Optimization of modulated fluid microchannels based on asymptotic homogenization". In: *Computer Methods in Applied Mechanics and Engineering* 419, p. 116646. DOI: [10.1016/j.cma.2023.116646](https://doi.org/10.1016/j.cma.2023.116646).
- Ferrari, Federico and Ole Sigmund (2019). "Revisiting topology optimization with buckling constraints". In: *Structural and Multidisciplinary Optimization* 59.5, pp. 1401–1415. DOI: [10.1007/s00158-019-02253-3](https://doi.org/10.1007/s00158-019-02253-3).
- Fuller, Thomas F and John N Harb (2018). *Electrochemical engineering*. John Wiley & Sons.
- Gersborg-Hansen, Allan et al. (2006). "Topology optimization of heat conduction problems using the finite volume method". In: *Structural and Multidisciplinary Optimization* 31.4, pp. 251–259.
- Giles, Michael B and Niles A Pierce (2000). "An introduction to the adjoint approach to design". In: *Flow, turbulence and combustion* 65.3, pp. 393–415. DOI: [10.1023/A:1011430410075](https://doi.org/10.1023/A:1011430410075).
- Giraldo, Sara et al. (2021). "Preparation of Composite Electrodes for All-Solid-State Batteries Based on Sulfide Electrolytes: An Electrochemical Point of View". In: *Batteries* 7.4. ISSN: 2313-0105. DOI: [10.3390/batteries7040077](https://doi.org/10.3390/batteries7040077).
- Goncalves, Juliano F et al. (2024). "Design of solid oxide fuel cell (SOFC) channel layout using the topology optimization method with a design variable propagation approach". In: *Structural and Multidisciplinary Optimization* 67.12, p. 216. DOI: [10.1007/s00158-024-03943-3](https://doi.org/10.1007/s00158-024-03943-3).
- Gowrisankaran, Gautam et al. (2016). "Intermittency and the value of renewable energy". In: *Journal of Political Economy* 124.4, pp. 1187–1234. DOI: [10.1086/686733](https://doi.org/10.1086/686733).
- Guo, Chao et al. (2022a). "Investigations on a novel cold plate achieved by topology optimization for lithium-ion batteries". In: *Energy* 261, p. 125097. ISSN: 0360-5442. DOI: [10.1016/j.energy.2022.125097](https://doi.org/10.1016/j.energy.2022.125097).
- Guo, Yong et al. (2022b). "Solid-state lithium batteries: Safety and prospects". In: *eScience* 2.2, pp. 138–163. ISSN: 2667-1417. DOI: [10.1016/j.esci.2022.02.008](https://doi.org/10.1016/j.esci.2022.02.008).
- Houser, Jacob et al. (2017). "Architecture for improved mass transport and system performance in redox flow batteries". In: *Journal of Power Sources* 351, pp. 96–105. ISSN: 0378-7753. DOI: [10.1016/j.jpowsour.2017.03.083](https://doi.org/10.1016/j.jpowsour.2017.03.083).

- Huang, Haocun et al. (2024). "Enhanced ion transport in ultrathin regenerated cellulose supercapacitor separators". In: *Journal of Materials Chemistry C* 12.25, p. 9189.
- Hvejsel, Christian Frier and Erik Lund (2011). "Material interpolation schemes for unified topology and multi-material optimization". In: *Structural and Multidisciplinary Optimization* 43, pp. 811–825.
- Ishida, Naoyuki et al. (2024). "Topology optimization for all-solid-state-batteries using homogenization method". In: *Structural and Multidisciplinary Optimization* 67.9, p. 162. DOI: [10.1007/s00158-024-03864-1](https://doi.org/10.1007/s00158-024-03864-1).
- Iwai, Hiroshi et al. (2011). "Power generation enhancement of solid oxide fuel cell by cathode/-electrolyte interface modification in mesoscale assisted by level set-based optimization calculation". In: *Journal of Power Sources* 196.7, pp. 3485–3495. ISSN: 0378-7753. DOI: [10.1016/j.jpowsour.2010.12.024](https://doi.org/10.1016/j.jpowsour.2010.12.024).
- Janek, Jürgen and Wolfgang G Zeier (2016). "A solid future for battery development". In: *Nature Energy* 1.9, pp. 1–4. DOI: [10.1038/nenergy.2016.141](https://doi.org/10.1038/nenergy.2016.141).
- Jensen, Jakob Søndergaard and Ole Sigmund (2011). "Topology optimization for nano-photonics". In: *Laser & Photonics Reviews* 5.2, pp. 308–321. DOI: [10.1002/lpor.201000014](https://doi.org/10.1002/lpor.201000014).
- Jin, Ning et al. (2018). "Parameter estimation of an electrochemistry-based lithium-ion battery model using a two-step procedure and a parameter sensitivity analysis". In: *International Journal of Energy Research* 42.7, pp. 2417–2430. DOI: [10.1002/er.4022](https://doi.org/10.1002/er.4022).
- Kambampati, Sandilya et al. (2021). "Level set topology optimization of load carrying battery packs". In: *International Journal of Heat and Mass Transfer* 177, p. 121570. ISSN: 0017-9310. DOI: [10.1016/j.ijheatmasstransfer.2021.121570](https://doi.org/10.1016/j.ijheatmasstransfer.2021.121570).
- KANNO, Ryoji (2023). "Developments in New Materials for Electrochemistry and Energy Storage Devices". In: *Electrochemistry* 91.10, pp. 102001–102001. DOI: [10.5796/electrochemistry.23-00064](https://doi.org/10.5796/electrochemistry.23-00064).
- Karaki, Hassan et al. (2023). "Optimizing the Microstructure and Processing Parameters for Lithium-Ion Battery Cathodes: A Use Case Scenario with a Digital Manufacturing Platform". In: *Energy Technology* 11.5, p. 2201032. DOI: [10.1002/ente.202201032](https://doi.org/10.1002/ente.202201032).
- Kawamoto, Atsushi et al. (2011). "Heaviside projection based topology optimization by a PDE-filtered scalar function". In: *Structural and Multidisciplinary Optimization* 44.1, pp. 19–24.
- Kench, Steve et al. (2024). "Li-ion battery design through microstructural optimization using generative AI". In: *Matter* 7.12, pp. 4260–4269. ISSN: 2590-2385. DOI: [10.1016/j.matt.2024.08.014](https://doi.org/10.1016/j.matt.2024.08.014).
- Khalik, Z. et al. (2021). "Parameter estimation of the Doyle – Fuller – Newman model for Lithium-ion batteries by parameter normalization, grouping, and sensitivity analysis". In: *Journal of Power Sources* 499, p. 229901. ISSN: 0378-7753. DOI: [10.1016/j.jpowsour.2021.229901](https://doi.org/10.1016/j.jpowsour.2021.229901).
- Kim, Patrick Joohyun H. et al. (2017). "Synergistic protective effect of a BN-carbon separator for highly stable lithium sulfur batteries". In: *NPG Asia Materials* 9.4, e375.
- Kim, Sangwook et al. (June 2022). "Projecting Recent Advancements in Battery Technology to Next-Generation Electric Vehicles". In: *Energy Technology* 10.8. DOI: [10.1002/ente.202200303](https://doi.org/10.1002/ente.202200303).

- Kimura, Yuta (2023). “Development of Measurement/Analysis Techniques to Bridge Microscopic and Macroscopic Electrochemical Phenomena in Energy Storage Devices”. In: *Electrochemistry* 91.10, pp. 101005–101005. DOI: [10.5796/electrochemistry.23-00074](https://doi.org/10.5796/electrochemistry.23-00074).
- Koç, Tuncay et al. (2022). “Toward optimization of the chemical/electrochemical compatibility of halide solid electrolytes in all-solid-state batteries”. In: *ACS Energy Letters* 7.9, pp. 2979–2987.
- Kondoh, Tsuguo et al. (2024). *Fundamentals of Topology Optimization: Applications to Elastic Structures and Thermal-fluid Related Engineering Problems (in Japanese)*. Corona Publishing Co., Ltd.
- Kozuka, Yuya et al. (2023). “Topology Optimization Based on the Level Set Method Considering Aesthetic Preference Using Texture Energy (in Japanese)”. In: *Transactions of the Japan Society of Mechanical Engineers* 89.924, pp. 22–00296. DOI: [10.1299/transjsme.22-00296](https://doi.org/10.1299/transjsme.22-00296).
- Lazard (2021). *Lazard’s Levelized Cost of Energy Analysis – Version 15.0*. <https://www.lazard.com/media/sptlfats/lazards-levelized-cost-of-energy-version-150-vf.pdf>. Accessed: 2025-08-09.
- Li, Hanyu et al. (2024). “Topology optimization for the full-cell design of porous electrodes in electrochemical energy storage devices”. In: *Structural and Multidisciplinary Optimization* 67.11, pp. 1–28.
- Li, Jinyao et al. (2023). “Improving the Electrochemical Performance of a Solid-State Battery with a LiFePO₄–Garnet-Based Composite Cathode”. In: *The Journal of Physical Chemistry C* 127.13, pp. 6192–6198.
- Lin, Tiras Y. et al. (2022). “Topology Optimization of 3D Flow Fields for Flow Batteries”. In: *Journal of The Electrochemical Society* 169.5, p. 050540. DOI: [10.1149/1945-7111/ac716d](https://doi.org/10.1149/1945-7111/ac716d).
- Lin, Tiras Y et al. (2024). “Shape matters: understanding the effect of electrode geometry on cell resistance and chemo-mechanical stress”. In: *Journal of the Electrochemical Society* 171.10, p. 100515.
- Liu, Fangfang and Xiuyun Chuan (2021). “Recent developments in natural mineral-based separators for lithium-ion batteries”. In: *RSC Advances* 11.27, p. 16633.
- Liu, L et al. (2012). “Modeling of solid oxide fuel cells with particle size and porosity grading in anode electrode”. In: *Fuel Cells* 12.1, pp. 97–108. DOI: [10.1002/fuce.201100095](https://doi.org/10.1002/fuce.201100095).
- Liu, Ping et al. (2009). “Design and fabrication of multifunctional structural batteries”. In: *Journal of Power Sources* 189.1. Selected Papers presented at the 14th INTERNATIONAL MEETING ON LITHIUM BATTERIES (IMLB-2008), pp. 646–650. ISSN: 0378-7753. DOI: [10.1016/j.jpowsour.2008.09.082](https://doi.org/10.1016/j.jpowsour.2008.09.082).
- Liu, S et al. (2015). *An identification method for enclosed voids restriction in manufacturability design for additive manufacturing structures*. DOI: [10.1007/s11465-015-0340-3](https://doi.org/10.1007/s11465-015-0340-3).
- Long, Mengly et al. (2024). “A method for estimating optimized porosity distribution in Reaction-Diffusion systems without reliance on topology optimization”. In: *Chemical Engineering Science* 298, p. 120420. ISSN: 0009-2509. DOI: [10.1016/j.ces.2024.120420](https://doi.org/10.1016/j.ces.2024.120420).
- Lu, Lirong et al. (2013). “Topology optimization of an acoustic metamaterial with negative bulk modulus using local resonance”. In: *Finite Elements in Analysis and Design* 72, pp. 1–12. ISSN: 0168-874X. DOI: [10.1016/j.finel.2013.04.005](https://doi.org/10.1016/j.finel.2013.04.005).

- Lv, Chade et al. (2022). "Machine learning: an advanced platform for materials development and state prediction in lithium-ion batteries". In: *Advanced Materials* 34.25, p. 2101474. DOI: [10.1002/adma.202101474](https://doi.org/10.1002/adma.202101474).
- Matsumori, Tadayoshi et al. (2013). "Topology optimization for fluid–thermal interaction problems under constant input power". In: *Structural and Multidisciplinary Optimization* 47, pp. 571–581. DOI: [10.1007/s00158-013-0887-8](https://doi.org/10.1007/s00158-013-0887-8).
- Messaggi, M. et al. (2018). "Analysis of flow field design on vanadium redox flow battery performance: Development of 3D computational fluid dynamic model and experimental validation". In: *Applied Energy* 228, pp. 1057–1070. ISSN: 0306-2619. DOI: [10.1016/j.apenergy.2018.06.148](https://doi.org/10.1016/j.apenergy.2018.06.148).
- Michell, A.G.M. (1904). "LVIII. The limits of economy of material in frame-structures". In: *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* 8.47, pp. 589–597. DOI: [10.1080/14786440409463229](https://doi.org/10.1080/14786440409463229).
- Minnmann, Philip et al. (2021). "Editors' choice—quantifying the impact of charge transport bottlenecks in composite cathodes of all-solid-state batteries". In: *Journal of The Electrochemical Society* 168.4, p. 040537.
- Mo, Xiaobao et al. (2021). "Topology optimization of cooling plates for battery thermal management". In: *International Journal of Heat and Mass Transfer* 178, p. 121612. ISSN: 0017-9310. DOI: [10.1016/j.ijheatmasstransfer.2021.121612](https://doi.org/10.1016/j.ijheatmasstransfer.2021.121612).
- Moës, Nicolas et al. (1999). "A finite element method for crack growth without remeshing". In: *International journal for numerical methods in engineering* 46.1, pp. 131–150.
- Multiphysics, COMSOL (1998). "Introduction to COMSOL multiphysics®". In: *COMSOL Multiphysics, Burlington, MA, accessed Feb 9, p. 2018*.
- Nagasubramanian, Ganesan and Kyle Fenton (2013). "Reducing Li-ion safety hazards through use of non-flammable solvents and recent work at Sandia National Laboratories". In: *Electrochimica Acta* 101, pp. 3–10. ISSN: 0013-4686. DOI: [10.1016/j.electacta.2012.09.065](https://doi.org/10.1016/j.electacta.2012.09.065).
- Nanda, Sanjay et al. (2021). "Anode-free full cells: a pathway to high-energy density lithium-metal batteries". In: *Advanced Energy Materials* 11.2, p. 2000804. DOI: [10.1002/aenm.202000804](https://doi.org/10.1002/aenm.202000804).
- Nelli Silva, Emílio C. et al. (1999). "Optimization methods applied to material and flextensional actuator design using the homogenization method". In: *Computer Methods in Applied Mechanics and Engineering* 172.1, pp. 241–271. ISSN: 0045-7825. DOI: [10.1016/S0045-7825\(98\)00231-X](https://doi.org/10.1016/S0045-7825(98)00231-X).
- Newman, J. and K.E. Thomas-Alyea (2004). *Electrochemical Systems*. The ECS Series of Texts and Monographs. Wiley. ISBN: 9780471477563.
- Noguchi, Y et al. (2015). "An acoustic metasurface design for wave motion conversion of longitudinal waves to transverse waves using topology optimization". In: *Applied Physics Letters* 107.22. DOI: [10.1063/1.4936997](https://doi.org/10.1063/1.4936997).
- Ong, Shyue Ping (2019). "Accelerating materials science with high-throughput computations and machine learning". In: *Computational Materials Science* 161, pp. 143–150. ISSN: 0927-0256. DOI: [10.1016/j.commatsci.2019.01.013](https://doi.org/10.1016/j.commatsci.2019.01.013).

- Osher, Stanley and James A Sethian (1988). "Fronts propagating with curvature-dependent speed: Algorithms based on Hamilton-Jacobi formulations". In: *Journal of Computational Physics* 79.1, pp. 12–49. ISSN: 0021-9991. DOI: [10.1016/0021-9991\(88\)90002-2](https://doi.org/10.1016/0021-9991(88)90002-2).
- Ouyang, Dongxu et al. (2019). "A review on the thermal hazards of the lithium-ion battery and the corresponding countermeasures". In: *Applied Sciences* 9.12, p. 2483. DOI: [10.3390/app9122483](https://doi.org/10.3390/app9122483).
- Pejman, Reza and Ahmad Raeisi Najafi (2023). "Multiphysics topology optimization of a multi-functional structural battery composite". In: *Structural and Multidisciplinary Optimization* 66.3, p. 46. DOI: [10.1007/s00158-023-03490-3](https://doi.org/10.1007/s00158-023-03490-3).
- Rao, Zhonghao and Shuangfeng Wang (2011). "A review of power battery thermal energy management". In: *Renewable and Sustainable Energy Reviews* 15.9, pp. 4554–4571. DOI: [10.1016/j.rser.2011.07.096](https://doi.org/10.1016/j.rser.2011.07.096).
- Reisacher, Elias et al. (2023). "Percolation Behavior of a Sulfide Electrolyte–Carbon Additive Matrix for Composite Cathodes in All-Solid-State Batteries". In: *Batteries* 9.12. ISSN: 2313-0105. DOI: [10.3390/batteries9120595](https://doi.org/10.3390/batteries9120595).
- Ren, Yaoyu et al. (2023). "Oxide-based solid-state batteries: a perspective on composite cathode architecture". In: *Advanced Energy Materials* 13.1, p. 2201939. DOI: [10.1002/aenm.202201939](https://doi.org/10.1002/aenm.202201939).
- Roy, Thomas et al. (2022). "Topology optimization for the design of porous electrodes". In: *Structural and Multidisciplinary Optimization* 65.6, p. 171.
- Ruan, Zhefei et al. (2022). "Incorporation of Poly(Ionic Liquid) with PVDF-HFP-Based Polymer Electrolyte for All-Solid-State Lithium-Ion Batteries". In: *Polymers* 14.10. ISSN: 2073-4360. DOI: [10.3390/polym14101950](https://doi.org/10.3390/polym14101950).
- Sarawutanukul, Sangchai et al. (2021). "Optimization of the electrode properties for high-performance Ni-rich Li-ion batteries". In: *ACS Applied Materials & Interfaces* 13.26, pp. 30643–30652. DOI: [10.1021/acsami.1c07019](https://doi.org/10.1021/acsami.1c07019).
- Sasaki, Takamitsu et al. (2024). "Topology optimization for 3D fluid diode design considering wall-connected structures". In: *Structural and Multidisciplinary Optimization* 67.12, p. 209. DOI: [10.1007/s00158-024-03920-w](https://doi.org/10.1007/s00158-024-03920-w).
- Schmidt, Oliver and Iain Staffell (Sept. 2023). "130Lifetime cost: Performing cost assessments". In: *Monetizing Energy Storage: A Toolkit to Assess Future Cost and Value*. Oxford University Press. ISBN: 9780192888174. DOI: [10.1093/oso/9780192888174.003.0005](https://doi.org/10.1093/oso/9780192888174.003.0005).
- Shahzad, Khurram et al. (2024). "Accelerating materials discovery: combinatorial synthesis, high-throughput characterization, and computational advances". In: *Science and Technology of Advanced Materials: Methods* 4.1, p. 2292486. DOI: [10.1080/27660400.2023.2292486](https://doi.org/10.1080/27660400.2023.2292486).
- Shi, Junli et al. (2015). "Porous membrane with high curvature, three-dimensional heat-resistance skeleton: a new and practical separator candidate for high safety lithium ion battery". In: *Scientific Reports* 5.1, p. 8255.
- Shi, Tan et al. (2020). "High active material loading in all-solid-state battery electrode via particle size optimization". In: *Advanced Energy Materials* 10.1, p. 1902881. DOI: [10.1002/aenm.201902881](https://doi.org/10.1002/aenm.201902881).

- Sigmund, O. and S. Torquato (1997). "Design of materials with extreme thermal expansion using a three-phase topology optimization method". In: *Journal of the Mechanics and Physics of Solids* 45.6, pp. 1037–1067. ISSN: 0022-5096. DOI: [10.1016/S0022-5096\(96\)00114-7](https://doi.org/10.1016/S0022-5096(96)00114-7).
- Sigmund, Ole and Jakob Søndergaard Jensen (2003). "Systematic design of phononic band-gap materials and structures by topology optimization". In: *Philosophical Transactions of the Royal Society of London. Series A: Mathematical, Physical and Engineering Sciences* 361.1806, pp. 1001–1019. DOI: [10.1098/rsta.2003.1177](https://doi.org/10.1098/rsta.2003.1177).
- Snyder, James F et al. (2007). "Electrochemical and mechanical behavior in mechanically robust solid polymer electrolytes for use in multifunctional structural batteries". In: *Chemistry of Materials* 19.15, pp. 3793–3801. DOI: [10.1021/cm070213o](https://doi.org/10.1021/cm070213o).
- Snyder, JF et al. (2008). "Multifunctional structural composite batteries for US army applications". In: *Proceedings of the 26th Army Science Conference*, pp. 1–8.
- Sung, Junghwan et al. (2024). "Recent advances in all-solid-state batteries for commercialization". In: *Materials Chemistry Frontiers* 8.8, pp. 1861–1887. DOI: [10.1039/D3QM01171B](https://doi.org/10.1039/D3QM01171B).
- Svanberg, Krister (1987). "The method of moving asymptotes—a new method for structural optimization". In: *International Journal for Numerical Methods in Engineering* 24.2, pp. 359–373. DOI: [10.1002/nme.1620240207](https://doi.org/10.1002/nme.1620240207).
- Takaoka, Kazuchiyo and Hiroshi Miyazu (2014). Pat. JP2014-86407A. Japan, in Japanese.
- Tian, Hong-Kang and Yue Qi (2017). "Simulation of the Effect of Contact Area Loss in All-Solid-State Li-Ion Batteries". In: *Journal of The Electrochemical Society* 164.11, E3512. DOI: [10.1149/2.0481711jes](https://doi.org/10.1149/2.0481711jes).
- Tjaden, Bernhard et al. (2016). "On the origin and application of the Bruggeman correlation for analysing transport phenomena in electrochemical systems". In: *Current Opinion in Chemical Engineering* 12. Nanotechnology / Separation Engineering, pp. 44–51. ISSN: 2211-3398. DOI: [10.1016/j.coche.2016.02.006](https://doi.org/10.1016/j.coche.2016.02.006).
- Tron, Artur et al. (2023). "Rational Optimization of Cathode Composites for Sulfide-Based All-Solid-State Batteries". In: *Nanomaterials* 13.2, p. 327.
- Wadbro, Eddie and Martin Berggren (2006). "Topology optimization of an acoustic horn". In: *Computer Methods in Applied Mechanics and Engineering* 196.1-3, pp. 420–436.
- Waki, Norihisa et al. (2012). Pat. JP2012-54114A. Japan, in Japanese.
- Wang, Michael Yu et al. (2003). "A level set method for structural topology optimization". In: *Computer Methods in Applied Mechanics and Engineering* 192.1-2, pp. 227–246.
- Wang, Renheng et al. (2020). "Lithium metal anodes: Present and future". In: *Journal of Energy Chemistry* 48, pp. 145–159. ISSN: 2095-4956. DOI: [10.1016/j.jechem.2019.12.024](https://doi.org/10.1016/j.jechem.2019.12.024).
- Wang, Sujun et al. (2025). "Multi-objective layout optimization of microstructures for redox flow battery using Galerkin decoupling technology and independent continuous mapping method". In: *Physics of Fluids* 37.7. DOI: [10.1063/5.0269715](https://doi.org/10.1063/5.0269715).
- Wei, Chaochao et al. (2023). "Dual N-modification enables high-performance Solid-State Li metal batteries with Li5.5PS4.5Cl1.5". In: *Chemical Engineering Journal* 476, p. 146531.

- Wolf, Amadeus et al. (2024). "Enhancing Flow Batteries: Topology Optimization of Electrode Porosity and Shape Optimization of Cell Design". In: *Energy Technology* 12.7, p. 2400244. DOI: [10.1002/ente.202400244](https://doi.org/10.1002/ente.202400244).
- Wong, EL et al. (2007). "Design and processing of structural composite batteries". In: *Proceedings of SAMPE*, pp. 3–7.
- Wu, Cong et al. (2022). "Solar power generation intermittency and aggregation". In: *Scientific reports* 12.1, p. 1363. DOI: [10.1038/s41598-022-05247-2](https://doi.org/10.1038/s41598-022-05247-2).
- Xu, Min et al. (2016). "Solid oxide fuel cell interconnect design optimization considering the thermal stresses". In: *Science Bulletin* 61.17, pp. 1333–1344. ISSN: 2095-9273. DOI: [10.1007/s11434-016-1146-3](https://doi.org/10.1007/s11434-016-1146-3).
- Xu, Q. et al. (2013). "Numerical investigations of flow field designs for vanadium redox flow batteries". In: *Applied Energy* 105, pp. 47–56. ISSN: 0306-2619. DOI: [10.1016/j.apenergy.2012.12.041](https://doi.org/10.1016/j.apenergy.2012.12.041).
- Yaji, Kentaro et al. (2018). "Topology optimization for the design of flow fields in a redox flow battery". In: *Structural and Multidisciplinary Optimization* 57, pp. 535–546. DOI: [10.1007/s00158-017-1763-8](https://doi.org/10.1007/s00158-017-1763-8).
- Yamada, Takayuki and Yuki Noguchi (2022). "Topology optimization with a closed cavity exclusion constraint for additive manufacturing based on the fictitious physical model approach". In: *Additive Manufacturing* 52, p. 102630.
- Yamada, Takayuki et al. (2010). "A topology optimization method based on the level set method incorporating a fictitious interface energy". In: *Computer Methods in Applied Mechanics and Engineering* 199.45, pp. 2876–2891. ISSN: 0045-7825. DOI: [10.1016/j.cma.2010.05.013](https://doi.org/10.1016/j.cma.2010.05.013).
- Yong-Gun, Lee et al. (2020). "High-energy long-cycling all-solid-state lithium metal batteries enabled by silver–carbon composite anodes". In: *Nature Energy* 5.4, pp. 299–308.
- Yoo, Jeonghoon and Noboru Kikuchi (2000). "Topology optimization in magnetic fields using the homogenization design method". In: *International Journal for Numerical Methods in Engineering* 48.10, pp. 1463–1479. DOI: [10.1002/1097-0207\(20000810\)48:10<1463::AID-NME952>3.0.CO;2-5](https://doi.org/10.1002/1097-0207(20000810)48:10<1463::AID-NME952>3.0.CO;2-5).
- Yoshino, Akira (2015). "Technical Trends of Separators for Lithium-ion Batteries (in Japanese)". In: *Journal of the Functional Paper Research Association* 59, pp. 9–13.
- Yuan, Hongyan et al. (2020). "Single lithium-ion conducting solid polymer electrolyte with superior electrochemical stability and interfacial compatibility for solid-state lithium metal batteries". In: *ACS Applied Materials & Interfaces* 12.6, pp. 7249–7256. DOI: [10.1021/acsami.9b20436](https://doi.org/10.1021/acsami.9b20436).
- Zhang, Heng et al. (2017). "Single lithium-ion conducting solid polymer electrolytes: advances and perspectives". In: *Chemical Society Reviews* 46.3, pp. 797–815. DOI: [10.1039/C6CS00491A](https://doi.org/10.1039/C6CS00491A).
- Zhang, Weihong et al. (2007). "Using strain energy-based prediction of effective elastic properties in topology optimization of material microstructures". In: *Acta Mechanica Sinica* 23.1, pp. 77–89.
- Zhang, Ying et al. (2020). "Towards better Li metal anodes: Challenges and strategies". In: *Materials Today* 33, pp. 56–74. ISSN: 1369-7021. DOI: [10.1016/j.mattod.2019.09.018](https://doi.org/10.1016/j.mattod.2019.09.018).

- Zhang, Z. et al. (2023a). "Review on composite solid electrolytes for solid-state lithium-ion batteries". In: *Materials Today Sustainability* 21, p. 100316. ISSN: 2589-2347. DOI: [10.1016/j.mtsust.2023.100316](https://doi.org/10.1016/j.mtsust.2023.100316).
- Zhang, Zhihua et al. (2023b). "Optimization of Annealing Process of Li₆PS₅Cl for All-Solid-State Lithium Batteries by Box–Behnken Design". In: *Batteries* 9.9, p. 480.
- Zhou, Yinghao et al. (2025). "Functional grading design of solid oxide fuel cell anodes: Insights from fabrication to operation simulations". In: *Chemical Engineering Journal* 520, p. 165908. ISSN: 1385-8947. DOI: [10.1016/j.cej.2025.165908](https://doi.org/10.1016/j.cej.2025.165908).
- Ziegler, Micah S and Jessika E Trancik (2021). "Re-examining rates of lithium-ion battery technology improvement and cost decline". In: *Energy & Environmental Science* 14.4, pp. 1635–1651. DOI: [10.1039/D0EE02681F](https://doi.org/10.1039/D0EE02681F).

About the Author

Ishida Naoyuki was born in Himeji, Hyogo, Japan, in March 1999.

In April 2017, after graduating from Hyogo Prefectural Himeji Nishi High School, he was enrolled in the Department of Physical Engineering, Faculty of Engineering at Kyoto University. In his sophomore year, he advanced to the Mechanical and Systems Engineering course, where he studied the four fundamental mechanics (Solid Mechanics, Fluid Dynamics, Thermodynamics, and Dynamics). For his final year project, he joined the Optimum Structure Design Engineering Laboratory under the supervision of Professor Shinji Nishiwaki, where he began his research on topology optimization.

He then advanced to the Graduate School of Engineering, Department of Mechanical Engineering and Science at Kyoto University through a five-year integrated program. In recognition of his academic excellence, he was awarded the Miura Award during his master's course. Following a change in laboratory affiliation to Strength of Materials and Structures Laboratory Department of Micro Engineering Graduate School of Engineering Kyoto University, he has been conducting his research under the supervision of Professor Kazuhiro Izui, focusing on the development of a topology optimization method for the design of all-solid-state batteries. He was also a recipient of Scholarship from DMG MORI CO., LTD for the 2023 and 2024 academic years. For the 2025 academic year, he was selected as a DC2 Research Fellow of the Japan Society for the Promotion of Science (JSPS).

After completing his doctoral course, he plans to continue his research as a postdoctoral researcher at Kyoto University. He intends to focus on the practical implementation of the optimization methods he has developed, aiming to translate his theoretical work into tangible applications.

December, 2025

Publication List

Peer-reviewed Journal Articles

1. **Ishida, N.**, Kondoh, T., Furuta, K., Li, H., Izui, K., and Nishiwaki, S., "Topology optimization for maximizing linear buckling load based on level set method." *Mechanical Engineering Journal* 9, no.4 (2022): 21-00425, DOI: [10.1299/mej.21-00425](https://doi.org/10.1299/mej.21-00425).
2. Hu, T., Wang, Y., Zhang, H., Li, H., **Ishida, N.**, Sasaki, T., Furuta, K., Izui, K., and Nishiwaki, S., "Multiscale topological design of coated structures with layer-wise bi-material lattice infill for minimum dynamic compliance." *Composites Structure* 323, (2023): 117468, DOI: [10.1016/j.compstruct.2023.117468](https://doi.org/10.1016/j.compstruct.2023.117468).
3. Soares da Costa Azevêdo, A., Li, H., **Ishida, N.**, Siqueira, L. O., Cortez, R. L., Nelli Silva, E. C., Nishiwaki, S., and Picelli, R., "Body-fitted topology optimization via integer linear programming using surface capturing techniques." *International Journal for Numerical Methods in Engineering* 125, no.13 (2024): e7480, DOI: [10.1002/nme.7480](https://doi.org/10.1002/nme.7480).
4. **Ishida, N.**, Furuta, K., Kishimoto, M., Hu, T., Iwai, H., Izui, K. and Nishiwaki, S., "Topology optimization for all-solid-state-batteries using homogenization method." *Structural and Multidisciplinary Optimization* 67, no.9 (2024): 162, DOI: [10.1007/s00158-024-03864-1](https://doi.org/10.1007/s00158-024-03864-1).
5. Furuta, K., Tsukuda, Y., Sasaki, T., **Ishida, N.**, Izui, K., Nishiwaki, S., and Watanabe, S., "Differential evolution-based topology optimization using Karhunen-Loève expansion." *Engineering Optimization*, (2024): 1–27, DOI: [10.1080/0305215X.2024.2400558](https://doi.org/10.1080/0305215X.2024.2400558).
6. Sasaki, T., Furuta, K., **Ishida, N.**, Li, H., Kondoh, T., Izui, K., and Nishiwaki, S., "Topology optimization for 3D fluid diode design considering wall-connected structures." *Structural and Multidisciplinary Optimization* 67, no. 12 (2024): 1–24, DOI: [10.1007/s00158-024-03920-w](https://doi.org/10.1007/s00158-024-03920-w).
7. 石田尚之, 佐々木貴光, 野口峻, 林翔太郎, 藤祥三, 古田幸三, 泉井一浩, 西脇眞二, "リチウムイオン電池用セパレータを対象としたトポロジー最適化." 精密工学会誌 91, 7 (2025): 791–800, DOI: [10.2493/jjspe.91.791](https://doi.org/10.2493/jjspe.91.791).
8. **Ishida, N.**, Furuta, K., Kishimoto, M., Sasaki, T., Iwai, H., Izui, K., and Nishiwaki, S., "Data-drive topology optimization of all-solid-state-batteries considering conductive additive material informed by microstructure analysis" *Structural and Multidisciplinary Optimization*,

68, no. 8 (2025): 164, DOI: [10.1007/s00158-025-04094-9](https://doi.org/10.1007/s00158-025-04094-9).

9. Hu, T., He, C., Yu, M., Nakayama, N., **Ishida, N.**, Kondoh, T., Furuta, K., Izui, K., and Nishiwaki, S., "Density-based topology optimization of coated structures with non-uniform coating thickness and smooth structural boundaries" *Structural and Multidisciplinary Optimization*, 68, no. 11 (2025): 224, DOI: [10.1007/s00158-025-04146-0](https://doi.org/10.1007/s00158-025-04146-0).

Conference Presentations

1. **Ishida, N.**, Kondoh, T., Furuta, K., Izui, K., and Nishiwaki, S., "Level Set-based Topology Optimization for Maximizing Linear Buckling Load." *14th World Congress of Structural and Multidisciplinary Optimization 2021 (WCSMO2021)*, Colorado, USA, June 13–18, 2021.
2. 石田尚之, 近藤継男, 古田幸三, 泉井一浩, 西脇眞二, "線形座屈荷重最大化を目的としたトポロジー最適化." 2021年度日本機械学会年次大会, 千葉, 日本, 2021年9月.
3. **Ishida, N.**, Li, H., Kondoh, T., Furuta, K., Izui, K., and Nishiwaki, S., "Topology optimization for a microfluidic mixing problem based on the level-set method." *Asian Congress of Structural and Multidisciplinary Optimization 2022 (ACSMO2022)*, Matsue, Japan, May 22–26, 2022.
4. **Ishida, N.**, Li, H., Kondoh, T., Furuta, K., Izui, K., and Nishiwaki, S., "Topology optimization Using Level-set Method for a Microfluidic Static Mixer." *Meshfree and Novel Finite Element Methods with Applications 2022 (MFEM2022)*, Berkeley, USA, September 25–27, 2022.
5. **Ishida, N.**, Li, H., Kondoh, T., Furuta, K., Izui, K., and Nishiwaki, S., "Level-set based topology optimization of a microfluidic mixing problem." *15th World Congress on Computational Mechanics & 8th Asian Pacific Congress on Computational Mechanics (WCCM-APCOM2022)*, Yokohama, Japan, July 31–August 5, 2022.
6. Li, H., Yu, M., Alexandersen, J., **Ishida, N.**, Kondoh, T., Furuta, K., Izui, K., and Nishiwaki, S., "Topology optimization of high resolution lattice structures using a PDE-filter." *32th Conference of Design & Systems*, Okayama, Japan, Sep 20–22, 2022.
7. 石田尚之, Li Hao, 近藤継男, 古田幸三, 泉井一浩, 西脇眞二, "静止型混合器を対象としたレベルセット法に基づくトポロジー最適化法の構築と流体モデルの比較." 2022年度日本機械学会年次大会, 富山, 日本, 2022年9月.
8. 尾関達大, 佐藤綾美, 石田尚之, 古田幸三, 泉井一浩, 西脇眞二, 茂木春樹, 島田貴弘, "熱流体問題を対象としたマルチスケルトポロジー最適化手法の開発." 第35回計算力学講演会(CMD2022), 鹿児島(オンライン), 日本, 2022年11月.
9. **Ishida, N.**, Kondoh, T., Kishimoto, M., Furuta, K., Izui, K., Iwai, H., and Nishiwaki, S., "Basic study of the structural optimization for all-solid-state batteries." *The 15th World Congress of Structural and Multidisciplinary Optimisation (WCSMO-15)*, Cork, Ireland, June 5–9, 2023.

10. **Ishida, N.**, Kondoh, T., Kishimoto, M., Furuta, K., Izui, K., Iwai, H., and Nishiwaki, S., "Fundamental Study of Topology Optimization for All-Solid-State Batteries." *The 17th U.S. National Congress on Computational Mechanics (USNCCM-17)*, Albuquerque, USA, July 23–27, 2023.
11. 石田尚之, 岸本将史, 佐々木貴光, 近藤継男, 古田幸三, 泉井一浩, 岩井裕, 西脇眞二, "全固体電池を対象としたトポロジー最適化に関する基礎検討." 2023年度日本機械学会年次大会, 東京, 日本, 2023年9月.
12. 佐々木貴光, 近藤継男, 石田尚之, Li Hao, 古田幸三, 泉井一浩, 西脇眞二, "浮遊固体領域を禁止する制約を用いた流体問題のトポロジー最適化." 2023年度日本機械学会年次大会, 東京, 日本, 2023年9月.
13. 鳥居悠吾, Sunghoon Lim, 石田尚之, 古田幸三, 泉井一浩, 西脇眞二, "熱損失を考慮した永久磁石モータの高効率設計のためのトポロジー最適化." 第33回設計工学・システム部門講演会, 日本機械学会, 金沢, 日本, 2023年9月.
14. 古田幸三, 佃泰聡, 石田尚之, 佐々木貴光, 泉井一浩, 西脇眞二, "メタマテリアル・メタサーフェス創出を目的とした差分進化型トポロジー最適設計法." 第33回設計工学・システム部門講演会, 日本機械学会, 金沢, 日本, 2023年9月.
15. 梅澤雄大, 石田尚之, Li Hao, 近藤継男, 古田幸三, Sunghoon Lim, 泉井一浩, 西脇眞二, "投影面積制約下の抗力最小化問題へのトポロジー最適化の適用." 第33回設計工学・システム部門講演会, 日本機械学会, 金沢, 日本, 2023年9月.
16. 佐々木貴光, 石田尚之, Li Hao, 古田幸三, 近藤継男, 泉井一浩, 西脇眞二, "流体機械を対象としたトポロジー最適化のための目的関数についての基礎検討." 日本機械学会第36回計算力学講演会(CMD2023), 愛知, 日本, 2023年10月.
17. Goda, H., **Ishida, N.**, Furuta, K., Izui, K., van Keulen, F., Kamei, K., Tsuchiya, T., Tabata, O., and Hirai, Y., "Electrode Design Using Multi-Material Topology Optimization for Accurate Measurement of Trans-Epithelial Electrical Resistance in Organ-on-a-Chip." *49th International Conference on Micro and Nano Engineering*.
18. **Ishida, N.**, Furuta, K., Kishimoto, M., Izui, K., Iwai, H., and Nishiwaki, S., "Topology optimization for all-solid-state-batteries considering a conductive additive." *The Asian Congress of Structural and Multidisciplinary Optimization 2024 (ACSMO 2024)*, Zhengzhou, China, May 19–23, 2024.
19. Noguchi, S., **Ishida, N.**, Sato, A., Motegi, H., Shimada, T., Nishiwaki, S., and Izui, K., "Functionally Graded Heat Sink Design Based on Data-Driven Multiscale Topology Optimization." *The Asian Congress of Structural and Multidisciplinary Optimization 2024 (ACSMO 2024)*, Zhengzhou, China, May 19–23, 2024.
20. **Ishida, N.**, Furuta, K., Kishimoto, M., Izui, K., and Nishiwaki, S., "Topology optimization for micro-structure of all-solid-state batteries." *9th European Congress on Computational Methods in Applied Sciences and Engineering (ECCOMAS 2024)*, Lisbon, Portugal, June

3–7, 2024.

21. 石田尚之, 古田幸三, 岸本将史, 岩井裕, 泉井一浩, 西脇眞二, "導電助剤を含む全固体電池を対象としたトポロジー最適化法の構築." 2024年度日本機械学会年次大会, 愛媛, 日本, 2024年9月.
22. 野口峻, 石田尚之, 佐藤綾美, 茂木春樹, 島田貴弘, 泉井一浩, 西脇眞二, 傾斜機能ヒートシンクのデータ駆動型最適設計法の構築, 2024年度日本機械学会年次大会, 2024年9月, 愛媛, 日本, J012p-10
23. 石田尚之, 佐々木貴光, 古田幸三, 岸本将史, 泉井一浩, 岩井裕, 西脇眞二, "ミクロ構造解析に基づき材料補間関数を用いた全固体電池のデータ駆動型トポロジー最適化." 日本機械学会第34回設計工学・システム部門講演会(D&S2024), 大阪, 日本, 2024年9月.
24. **Ishida, N.**, Furuta, K., Izui, K., and Nishiwaki, S., "On the topology optimization for all-solid-state battery designs considering charge-discharge processes." *The 16th World Congress of Structural and Multidisciplinary Optimization (WCSMO-16)*, Kobe, Japan, May 18–23, 2025.
25. Otomori, M., Li, H., **Ishida, N.**, Izui, K., and Nishiwaki, S., "Topology optimization for the design of insulating shield in electroplating process using tertiary current distribution." *The 16th World Congress of Structural and Multidisciplinary Optimization (WCSMO-16)*, Kobe, Japan, May 18–23, 2025.
26. Noguchi, S., **Ishida, N.**, Sato, A., Motegi, H., Shimada, T., Nishiwaki, S., and Izui, K., "On the topology optimization for functionally graded heat sinks with plate fins." *The 16th World Congress of Structural and Multidisciplinary Optimization (WCSMO-16)*, Kobe, Japan, May 18–23, 2025.
27. Siqueira, L., Noguchi, S., **Ishida, N.**, Silva, E., Nishiwaki, S., and Picelli, R., "Topology Optimization of transient fluid-structure interaction problems considering binary design variables and explicit smooth boundaries." *The 16th World Congress of Structural and Multidisciplinary Optimization (WCSMO-16)*, Kobe, Japan, May 18–23, 2025.
28. 石田尚之, 古田幸三, 泉井一浩, 西脇眞二, 充放電を考慮する全固体電池を対象としたトポロジー最適化法の構築, 第30回計算工学講演会, 2025年6月, 埼玉, 日本, A-04-05
29. 野口峻, 石田尚之, 佐藤綾美, 茂木春樹, 島田貴弘, 西脇眞二, 泉井一浩, 傾斜機能ヒートシンクのトポロジー最適化及び設計形状の生成, 第30回計算工学講演会, 2025年6月, 埼玉, 日本, A-06-03
30. 石田尚之, 古田幸三, 泉井一浩, 西脇眞二, 異方性材料からなる全固体電池を対象としたトポロジー最適化, 第35回設計工学・システム部門講演会, 2025年9月, 東京, 日本, 2215
31. 石田尚之, 古田幸三, 泉井一浩, 西脇眞二, 複数回の充放電を考慮する全固体電池を対象としたトポロジー最適化, 2025年度日本機械学会年次大会, 2025年9月, 北海道, 日本, J121-05