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TOPICAL REVIEW

Recent advances in electrode optimization of electrochemical energy devices using topology optimization

Mehrzad Alizadeh[1,3], **Patcharawat Charoen-Amornkitt**[2,3], **Takahiro Suzuki**[1] **and Shohji Tsushima**[1,*]

[1] Department of Mechanical Engineering, Graduate School of Engineering, Osaka University, Suita, Osaka 565-0871, Japan
[2] Electrochemical Energy Storage and Conversion Laboratory, Department of Mechanical Engineering, Faculty of Engineering, King Mongkut's University of Technology Thonburi, 126 Pracha Uthit Rd., Bang Mod, Thung Khru, Bangkok 10140, Thailand
[3] These authors contributed equally to the study and should be considered the co-first authors.
[*] Author to whom any correspondence should be addressed.

E-mail: tsushima@mech.eng.osaka-u.ac.jp

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Abstract

Topology optimization (TO) has emerged as a prominent trend in recent years, driven by its ability to explore optimized material distributions from scratch. Recently, there has been a significant shift in the application of TO, in optimizing systems involving complex electrochemical reactions, particularly electrode porous structures. This paper aims to examine the utilization of TO in enhancing electrodes across various electrochemical energy devices (EEDs). It encompasses a broad spectrum of applications, including the optimization of porous electrodes through the density-based method and interfaces between electrodes and electrolytes through the level-set method. The paper will delve into the challenges and opportunities associated with employing TO in electrode design for EEDs. These challenges involve addressing computational complexity, the absence of theoretical foundations for optimized structures, and the fabrication of complex structures for practical real-world applications. Additionally, beyond TO, the paper will spotlight other notable techniques in the structural design of porous electrodes using mathematical optimization. By offering insights into state-of-the-art research and developments in TO's application to electrode design, this paper provides researchers with valuable resources to navigate the evolving landscape of electrode design for EEDs.

1. Introduction

With energy production being a major contributor to greenhouse gas emissions, electrochemical energy storage and conversion systems are critical components in the global shift towards renewable energy adoption. One of the primary challenges in this transition stems from the intermittent nature of renewable energy sources, such as solar and wind power. While electricity can be generated efficiently from these sustainable resources, its production is subject to variations in weather conditions and daylight hours. Consequently, this intermittent availability poses limitations and uncertainties on a continuous and stable supply of energy. To address this challenge, electrochemical energy storage and conversion systems serve as vital solutions. These systems enable the efficient storage of excess energy generated during periods of high production, subsequently distributing it during times of low generation or high demand. The incorporation of electrochemical energy devices (EEDs), such as batteries and fuel cells, can enhance the reliability of renewable energy sources by mitigating the dependence on external factors like weather patterns. This improvement can take various forms, including but not limited to the storage of electricity through secondary batteries, the conversion of chemical energy stored in molecular bonds into electricity, as observed in fuel cells, or the utilization of surplus electricity to produce energy carrier substances and other valuable fuels or compounds, such as through water or carbon dioxide electrolyzers. In essence, these systems provide a means to bridge the gap between energy supply and demand, thereby facilitating the integration of renewable energy into the mainstream power grid. In addition to stationary applications, certain EEDs,

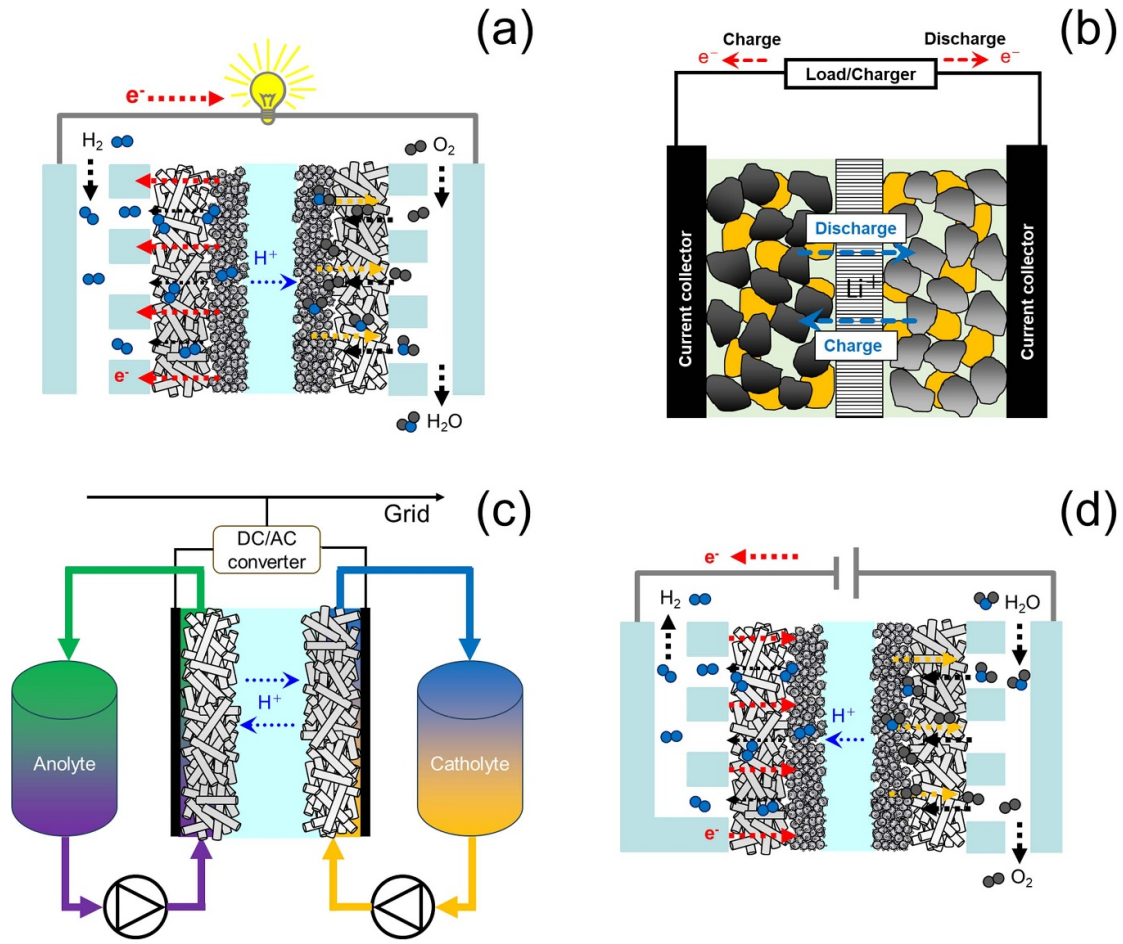

Figure 1. Schematic diagram showing the components of EEDs including (a) polymer electrolyte fuel cells, (b) lithium-ion batteries, (c) redox flow batteries, and (d) proton exchange membrane water electrolyzers.

exemplified by lithium-ion batteries, have found increasing utility in the transportation sector, particularly in electric vehicles. As a result, electrochemical energy storage and conversion technologies play a pivotal role in enabling a sustainable and resilient energy future.

EEDs comprise various components that depend on their specific type and application. However, they all share a critical component, the 'electrode', which typically functions as a porous medium composed of one or several materials. Schematics of various EEDs with their components are shown in figure 1. The performance and lifespan challenges in EEDs are particularly associated with their electrodes, where essential electrochemical reactions occur alongside various transport phenomena. These phenomena include the transfer of mass, momentum, heat, and charge, all of which are critical for the device's overall performance. Electrodes serve as the interfaces where electrochemical reactions take place, converting chemical energy into electrical energy (and vice versa) or breaking chemical bonds using electricity. However, achieving optimal electrode performance requires comprehensive understanding of complex interactions between the materials used, the design of the electrode structure as well as other components, and the dynamics of transport and rate processes within the device. To drive widespread adoption of EEDs in commercial applications, several key objectives must be addressed. First, there is a need to reduce the fabrication costs associated with electrode materials and manufacturing processes. This involves finding cost-effective materials and production methods without compromising performance or durability. Furthermore, there is a continuous push to enhance the overall performance of EEDs. This includes increasing energy efficiency and capacity, power output, and stability while minimizing losses and degradation over time. Improving electrode design and optimizing material properties are crucial aspects of achieving these performance enhancement requirements. Lastly, extending the operational lifespan of EEDs is paramount for their practical utility and economic viability. This involves developing electrode materials and configurations that can withstand prolonged operation under various operating conditions without significant degradation or loss of performance. Addressing the challenges associated with electrodes in EEDs requires a multidisciplinary approach that integrates materials science, engineering, and electrochemistry.

Given the pivotal role of electrodes in these devices, one of the key challenges in improving electrode performance lies in minimizing irreversible losses that are attributed to the transport phenomena and electrochemical reactions. Irreversible losses can result from various factors, including overpotentials, electrical resistance in the electrode materials, sluggish mass transport, and side reactions that consume energy without contributing to the desired output. To address these challenges and improve electrode performance, a direct approach involves redesigning the electrode structure to minimize these irreversible losses. This can include optimizing the material, composition, and morphology of the electrode to enhance their electrochemical activity and transport properties while mitigating undesirable side reactions and other phenomena. By focusing on minimizing irreversible losses in electrodes, researchers aim to maximize the efficiency and overall performance of EEDs.

In the quest to enhance the performance of EEDs, researchers have explored various avenues, including the synthesis of novel materials [1–3] and modification of electrodes [4–7]. However, despite these strides, many previous studies [8–10] have relied on a trial-and-error approach to optimize electrode performance. In this methodology, researchers systematically test different materials, configurations, or fabrication techniques to identify optimal conditions for device operation. While this approach has yielded valuable insights and incremental improvements, it can be time-consuming, resource-intensive, and limited in its ability to explore the full design space for a broad range of applications and operational conditions. Moreover, this approach primarily relies on the researcher's intuition and experience, potentially limiting the exploration of design solutions that may not be readily realizable. As a result, there is a growing recognition of the need for more systematic and efficient approaches to electrode design and optimization. By leveraging computational modeling and simulation, advanced characterization techniques, and optimization algorithms, researchers aim to accelerate the discovery and development of high-performance electrode materials and structures. These approaches allow for a more comprehensive exploration of the design space, enabling researchers to identify optimal design solutions more effectively while minimizing the need for extensive experimental testing. Furthermore, by integrating computational modeling with experimental validation, researchers can gain deeper insights into the underlying mechanisms governing multiphysics phenomena occurring in electrodes at nano to macro-scales. This synergistic approach enables researchers to develop a more fundamental understanding of electrochemical processes and design principles, leading to the development of next-generation EEDs with enhanced performance and functionality.

Topology optimization (TO) has emerged as a systematic bottom–up design approach in material informatics, allowing for the spatial redistribution of materials to achieve enhanced structural performance within a specified design domain [11]. It is defined as a computational method for optimizing material distribution within a defined design space to achieve the best possible performance while meeting specific constraints. While TO has been successfully applied to address various physical challenges in engineering and design [12–18], its application in optimizing systems involving complex (electro-) chemical reactions, particularly in the realm of electrochemistry, has presented ongoing challenges. In the domain of EEDs, TO has previously been utilized for optimizing the design of components such as flow channels [19–21], cooling plates [22–25], and end plates [26–29]. However, recent years have witnessed a notable shift in focus towards the application of TO in the design of porous electrodes across various EEDs. The adoption of TO for porous electrode design represents a departure from traditional approaches and opens up new possibilities for enhancing the performance of EEDs that was not possible through conventional methods. By systematically reshaping the microstructure of porous electrodes, TO offers the potential to optimize key transport and rate properties such as effective mass diffusivity, charge conductivity, and electrochemical reactive surface area. This shift in approach reflects a growing recognition of the importance of electrode design in determining overall device performance and efficiency. The adoption of TO in porous electrode design represents a promising avenue for advancing the field of electrochemical energy conversion and storage. By leveraging the principles of material informatics and systematic design optimization, researchers aim to unlock new insights into the fundamental relationships between electrode microstructure, electrochemical performance, and device efficiency.

This review seeks to provide a comprehensive examination of the utilization of TO in the design and enhancement of electrodes across various EEDs such as secondary batteries, fuel cells, electrolyzers, and capacitors. These devices are pivotal in the storage, conversion, and utilization of energy. Throughout the article, as outlined in figure 2, we will delve into diverse applications of TO in electrode design, encompassing areas like the optimization of porous electrodes and the interfaces between electrodes and electrolytes. Additionally, the present review will explore the challenges and opportunities associated with applying TO in EEDs electrode design. These challenges include managing computational complexity, addressing the lack of theoretical underpinning for optimized structures, and overcoming obstacles in fabricating complex structures for real-world practical applications. Moreover, beyond TO, we will also highlight some other noteworthy techniques in the structural design of porous electrodes based on mathematical optimization.

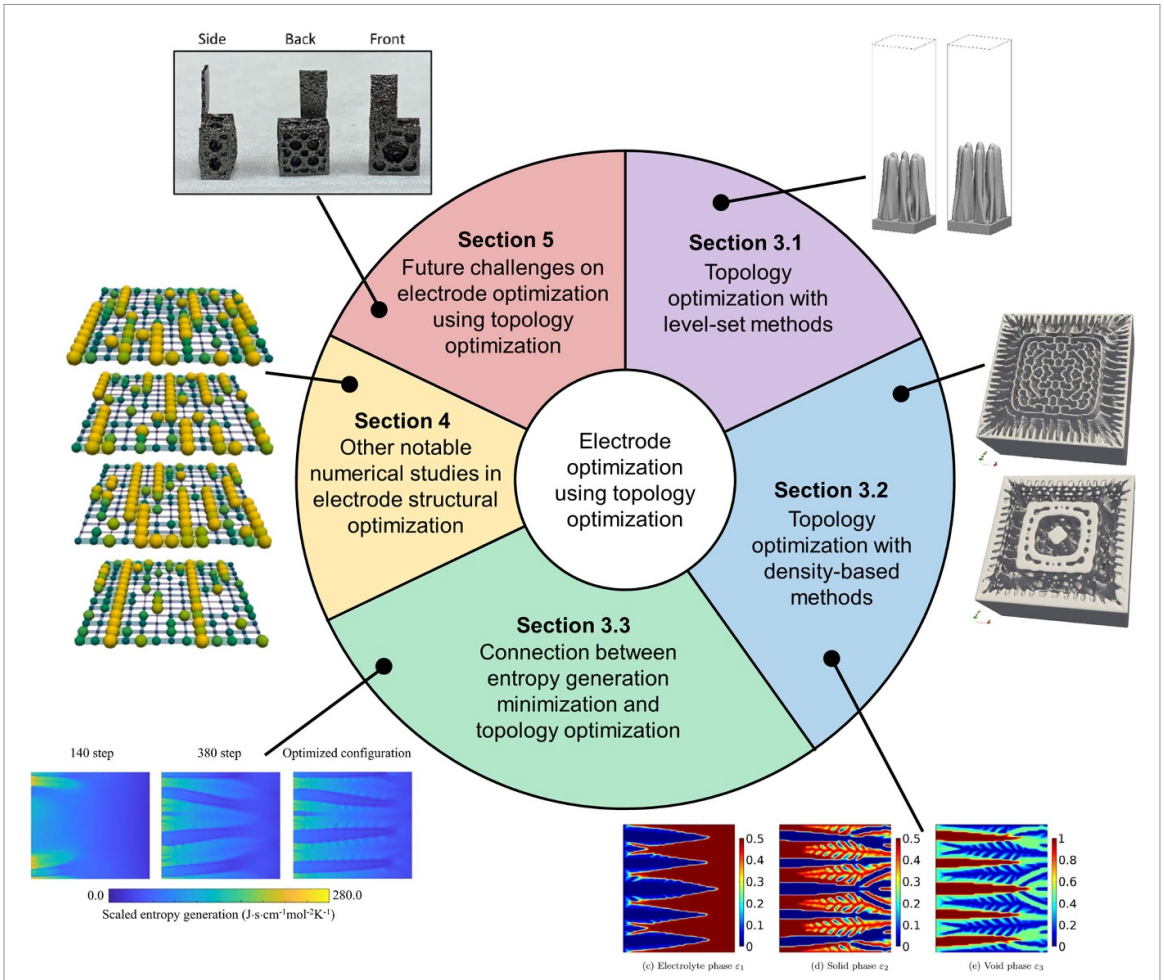

Figure 2. An illustration summarizing the organization of this review (Reproduced from [30], Copyright (2019), with permission from IOP Publishing; Reproduced from [31], with permission from Springer Nature; Reprinted from [32], Copyright (2023), with permission from Elsevier; Reprinted from [33], Copyright (2023), with permission from Elsevier; Reproduced from [34]. CC BY 4.0; Reprinted from [35], Copyright (2023), with permission from Elsevier).

The aim is to offer insights into cutting-edge research and developments in TO's application to electrode design, providing researchers with valuable resources to navigate the evolving landscape of EEDs.

2. Topology optimization

TO is a computational technique employed for the automated creation of an optimal structural layout, achieved by identifying the most efficient spatial allocation of material across a predetermined design space [11, 12]. The primary objective of TO is to enhance the performance of a structure, adhering to prescribed design specifications and constraints. Though both fall under the umbrella of layout optimization, TO adopts a more radical approach compared to shape optimization. Unlike shape optimization, which refines an existing design by adjusting its boundaries while preserving the overall layout, TO transcends this limitation. It treats the design space as a vast, unexplored territory, employing computational algorithms to identify the optimal layout from scratch [36]. This allows TO to potentially discover entirely new configurations that may not have been conceived with traditional approaches. Essentially, TO entails iteratively modifying the material allocation within a designated design space to optimize structural performance. Through the elimination of material from non-essential zones and its redistribution to critical areas, the design is refined to fulfill performance goals, such as weight reduction, stiffness maximization, or stress concentration minimization. In the field of porous electrodes, these goals may extend to other factors beyond mechanical properties. These factors include but are not limited to high reactive surface area, electrical charge conductivity, and hydraulic permeability, which ultimately contribute to the overall performance of EEDs.

TO originates from structural engineering and computational mechanics. The concept of enhancing structural efficacy by optimizing material distribution emerged in the latter part of the 20th century [12]. An important contribution to contemporary TO methodologies is evident in the work of Bendsøe and Kikuchi, documented in their publication of 1988 [37]. In this paper, Bendsøe and Kikuchi pioneered the application

of homogenization techniques to generate optimal structural topologies. This work represented a notable progression in structural optimization, showcasing the viability of optimizing material configurations to attain enhanced structural integrity. Building on this foundational work, subsequent researchers advanced the theoretical framework, methodological approaches, and application domains of TO. The integration of mathematical optimization techniques, sophisticated algorithms, and computational advancements empowered engineers and designers to explore unconventional material distributions and generate high-performing structural designs. Consequently, the application of TO has proliferated across diverse problems, such as mechanical [12], chemical [33], thermal [13, 38], fluid [16], microfluidics [36], and acoustic [39] systems. The potential of TO to automate design processes and generate structurally efficient solutions has established it as an indispensable tool for engineers seeking to optimize performance through improved structural designs. As research efforts and computational methods continue to mature, the field of TO undergoes continuous evolution, unveiling novel avenues for structural optimization and fostering advancements in structural efficiency and design innovation.

The field of TO has witnessed significant advancements, with researchers introducing diverse strategies for optimizing structural configurations. Existing literature can be categorized based on various criteria, including parameterization methods, system modeling, optimization algorithms, and design update schemes, each offering advantages and drawbacks. Given the wide variety of TO approaches, we briefly introduce the main techniques used for electrode design. Interested readers may refer to previously published comprehensive reviews [11, 12, 40–43] for more details on mathematical and algorithmic foundations of TO. It is noteworthy that these reviews primarily focus on compliance optimization in mechanical design problems, such as the classical Messerschmitt–Bölkow–Blohm beam and cantilever beam. In contrast, optimizing electrode structures involves multi-physics systems that integrate various physical and chemical phenomena, presenting challenges beyond the mathematical aspects of TO, which is the focus of the present paper. Selecting the appropriate TO framework hinges on factors like problem formulation, geometric complexity, relevant physical phenomena, and chosen models for performance evaluation. Design parameterization refers to the method used to establish the connection between design variables and the resulting physical properties [44]. Two dominant approaches include density-based and level-set methods. Bendsøe and Kikuchi [37] introduced the concept of numerical homogenization, aiming to modify the internal topology for achieving anisotropic material properties instead of solely focusing on boundary variations. This approach leverages homogenization theory, which estimates the effective material properties at a macroscopic level by treating the material as a uniform medium [12]. It is important to note, as pointed out in [12], that the terms 'micro' and 'macro' scales within this context are used comparatively and do not correspond to specific length scales (such as 'microscale' or 'microstructure' typically referring to sizes less than 1 mm). Following homogenization, a density-based method represented by solid isotropic material with penalization (SIMP) was introduced [45]. Density-based TO is a technique that modifies the density of elements within a fixed finite element mesh, using an interpolation function to adjust mechanical properties and determine the optimal distribution of solid and void material. SIMP, the most commonly used version of density-based TO, seeks to find the optimal material distribution based on an auxiliary density function assigned to each discrete element of the design domain, with values ranging from zero to one. Here, a density $\rho = 1$ and $\rho = 0$ represent solid and void phases, respectively. Intermediate density values ($0 < \rho < 1$) correspond to fictitious materials with properties (e.g. Young's modulus in mechanics or thermal conductivity in heat transfer) lying between those of solid and void phases. While these 'gray elements' lack an explicit physical interpretation, the introduction of a continuous density function significantly simplifies the mathematical calculations by transforming the problem from an integer-based to a continuous formulation. It is crucial to remember that the material properties within each element are assumed to be homogeneous, even though the overall (global) material behavior exhibits heterogeneity throughout the entire design domain due to the varying density distribution. Although the properties of the solid and void phases are known, SIMP interpolation, i.e. a power-law relationship, is employed to estimate the properties of these intermediate-density elements. The penalized material density can then be defined as:

$$\rho_{\text{penalized}} = \rho_{\min} + (1 - \rho_{\min})\,\rho^{p} \tag{1}$$

where $\rho_{\text{penalized}}$ is the penalized density, $\rho_{\min}$ is minimum penalized density, and p is a penalty exponent. It is noteworthy that while the penalized density can theoretically reach zero, introducing a minimum penalized density is often necessary for numerical stability during optimization. The penalty exponent, p, plays a crucial role in steering the optimizer towards assigning either solid or void densities ($\rho = 0$ or 1). By increasing the cost associated with intermediate densities, the penalty term discourages the formation of 'gray elements.' In other words, assigning a partial density leads to higher material usage without a significant improvement in beneficial properties like mechanical stiffness. Consequently, the optimizer prioritizes assigning densities

close to either zero or one, ultimately leading to a clear distinction between solid and void regions within the design. The effective material properties in each element as a function of penalized density is given by [44]:

$$\lambda^{\text{eff}} = \lambda_{\min} + (\lambda_{\max} - \lambda_{\min})\,\rho_{\text{penalized}} \tag{2}$$

in which $\lambda_{\min}$ and $\lambda_{\max}$ are minimum and maximum values of physical properties, corresponding to the void and solid phases. Other interpolation schemes with similar principles exist, such as rational approximation of material properties and Darcy [46–48]. Porous electrodes in EEDs typically comprise multiple phases. For instance, carbon fiber electrodes contain a carbon phase and voids, while fuel cell catalyst layers involve a mix of catalyst material, support material, polymer binder, and voids. Extensive research has explored the connection between electrode microstructure and key properties like catalytic activity, mass diffusivity, reactive surface area, and permeability. These studies employ experimental, numerical, and theoretical approaches to establish correlations linking macroscopic material characteristics (e.g. volume fraction of each phase or porosity) to bulk material properties [49–58]. A prominent example is the Bruggeman equation, which relates tortuosity and porosity based on effective medium theory [52]. Despite their limitations, assuming these established relationships hold true at the microscopic (element) level, and considering that each element is isotropic and homogeneous, existing correlations could be leveraged for topological optimization of electrode microstructure instead of SIMP.

While density-based TO is popular due to its broad applicability and ability to avoid re-meshing during optimization, it struggles to capture intricate interface shapes between different phases. Level-set parameterization methods address this limitation by representing the structure's geometry with a level-set function. Unlike traditional explicit boundary representations, where the geometry is explicitly defined, the level-set method utilizes a higher-dimensional function, the level set function. This scalar function mathematically defines the interface between various materials or phases within the design space [59]. Each point in the design domain receives a value from the level-set function, with positive values indicating one material (e.g. solid), negative values indicating another (e.g. void), and zero representing the exact location of the interface, as expressed by:

$$\begin{cases} \Phi(\mathbf{x}) > 0; & \mathbf{x} \in \Omega \\ \Phi(\mathbf{x}) = 0; & \mathbf{x} \in \partial\Omega \\ \Phi(\mathbf{x}) < 0; & \mathbf{x} \in D \backslash \Omega \end{cases} \tag{3}$$

where Φ is the level-set function, $\mathbf{x}$ represent any point in the design domain, and D, Ω, and $\partial\Omega$ are the design domain, material domain, and the interface between the two phases, respectively. By cleverly manipulating this level set function, the method can track the interface's motion implicitly. This method excels at capturing sharp boundaries and complex interfaces between materials, making it ideal for problems in which performance hinges on interfacial properties or behavior. This approach offers significant advantages by decoupling the representation of the geometry from its topology, allowing for seamless handling of complex topological changes such as merging, splitting, or evolving boundaries [60]. Moreover, the use of an implicit representation allows for easy incorporation of shape optimization techniques, offering designers and engineers a versatile toolset for achieving optimal geometric configurations tailored to specific design objectives [59]. However, the final design obtained from this method can be significantly influenced by the initial configuration provided [44]. The level set method's inherent numerical stability and avoidance of mesh-dependent spatial oscillations, such as staircasing, further enhance its applicability in diverse engineering domains [59]. Regardless of the chosen parameterization method, all TO procedures rely on optimization algorithms to update design solutions. Updating design solutions involve evolution of either density or level-set function through an algorithmic procedure. These algorithms can be gradient-based, such as Method of Steepest Descent [61], sequential linear programming [62], method of moving asymptotes (MMA) [63], globally convergent MMA [64]. Alternatively, derivative-free methods [65], such as evolutionary algorithms [66] can be employed. While evolutionary structural optimization (ESO) methods are not the focus of this review, it is worth mentioning that these approaches are often computationally expensive and may get stuck in local optima.

Gradient-guided TO involves an iterative process, akin to other optimizations. The optimization process begins with defining the problem and formulating the objective function. The primary optimization loop then starts with an initialization step, where an initial design is created. Subsequently, the objective function value is assessed, typically as a function of one or multiple state variables, necessitating the solution of a set of governing equations constituting the mathematical model. Researchers employ various modeling and simulation techniques to describe system behavior, such as the finite element method, finite volume method, and lattice Boltzmann method (LBM). Upon solving the state equations and obtaining the values of state variables, the objective function is determined. Next, a sensitivity analysis is performed to evaluate the

gradient of the objective function with respect to the design variables (e.g. material densities), often employing variational calculus methods such as the adjoint state method [67]. Subsequent to sensitivity analysis, regularization techniques [59, 68] may be employed to address issues such as the checkerboard pattern problem [11, 69, 70], eliminate numerical artifacts, and to promote convergence and smooth solutions. The design variables are then updated utilizing the optimizer and the sensitivity information. This iterative process continues until a convergence criterion, such as a maximum number of iterations, is met. It is worth noting that, as with any optimization problem, the initial configuration (initialization step of optimization) and model boundary conditions play a critical role in shaping the optimization process. An unsuitable initialization may cause the method to converge slowly or get trapped in a local optima. Similarly, improper boundary conditions can lead to unrealistic or unphysical outcomes. To minimize these risks, sensitivity analysis can be conducted to assess how different initial configurations and boundary conditions influence the results. This involves running the optimization with various initial shapes (layouts) and boundary conditions to observe their impact on the final solution. Such analysis helps ensure that the results are robust and not overly dependent on arbitrary choices of conditions.

Although not within the scope of this paper, other TO approaches are worth noting, including ESO [71], bi-directional ESO (BESO) [71], moving morphable component (MMC) [72], and moving morphable void (MMV) [73]. ESO optimizes structures by progressively removing elements with the least stress. BESO extends this by also adding new elements in high-stress areas, allowing for improvements from both directions. MMC and MMV, based on explicit Lagrangian descriptions, are dual methods. MMC uses adaptable components that can move, change shape, overlap, and merge, facilitating precise geometric designs and complex structures. In contrast, MMV employs voids to refine the topology. Both methods use geometric approaches to optimization, reviving classical shape optimization techniques.

3. Topology optimization for electrodes of EEDs

In the preceding section, we discussed two primary methodologies employed in TO, namely level-set and density-based approaches. In the realm of porous electrode design, level-set methods have been prevalent in studies examining the influence of interfacial boundaries between different phases (e.g. electrode–electrolyte interface shape). In this case, interfaces between material phases are defined implicitly by iso-contours of a level-set function. This implicit function provides a clear description of the boundaries, enhancing the accuracy of the response captured near the boundaries and eliminating ambiguities associated with intermediate material phases encountered in density-based approaches. Consequently, the chosen mathematical model should be capable of capturing the specific phenomena under investigation, particularly how structural changes impact those phenomena. Density-based methodologies, on the other hand, find application in scenarios where optimization of macroscopic properties distribution—such as porosity or solid volume fraction—is the focal point. As previously mentioned, density-based methods describe the layout through a set of material distribution functions, comprising two or more phases, with one phase typically representing 'no material' (i.e. the void phase). This material distribution is often discretized using element-wise constant or nodal shape functions. The following subsections review previous works that utilize these two approaches for structural design of porous electrodes in EEDs.

3.1. Level-set methods

The initial investigations in the area of electrode optimization focused on employing structural TO using level-set methods. This can be traced back to 2011 (see figures 3(a)–(c)) when Iwai *et al* [74] conducted optimization based on level-set techniques to explore the optimized cathode-electrolyte interface of solid oxide fuel cells (SOFCs) at meso-scale. The authors used a 2D model of a SOFC to find the best design for maximizing current density at a fixed voltage level. Their simulation included the entire cell: the anode, electrolyte, and cathode. These components were 300 microns long (through-plane direction) and 50 microns wide (in-plane direction). However, they only optimized the design of a smaller rectangular area within the cathode and electrolyte, measuring 150 microns long by 50 microns wide. They discovered that a non-flat wavy design of the cathode-electrolyte interface led to improved performance compared to the conventional flat interface as seen in figure 3(a). This figure depicts the changes of interface shape throughout the optimization process. As shown in figure 3(a), the initial interface had a step-like rectangular shape; however, the optimizer favored a more curved form in the final step. The optimal interface shape, however, depends on the simulation conditions. Their findings suggest that a flatter interface is preferable when gas diffusion resistance within the cathode is higher. Although the fabricated interface did not precisely match the optimized design obtained from mathematical optimization (see figure 3(b)), preliminary experiments involving the modification of the interface, such as fabricating grooved electrodes (a structure similar to the optimized results to some extent), demonstrated enhanced performance compared to electrodes with a flat

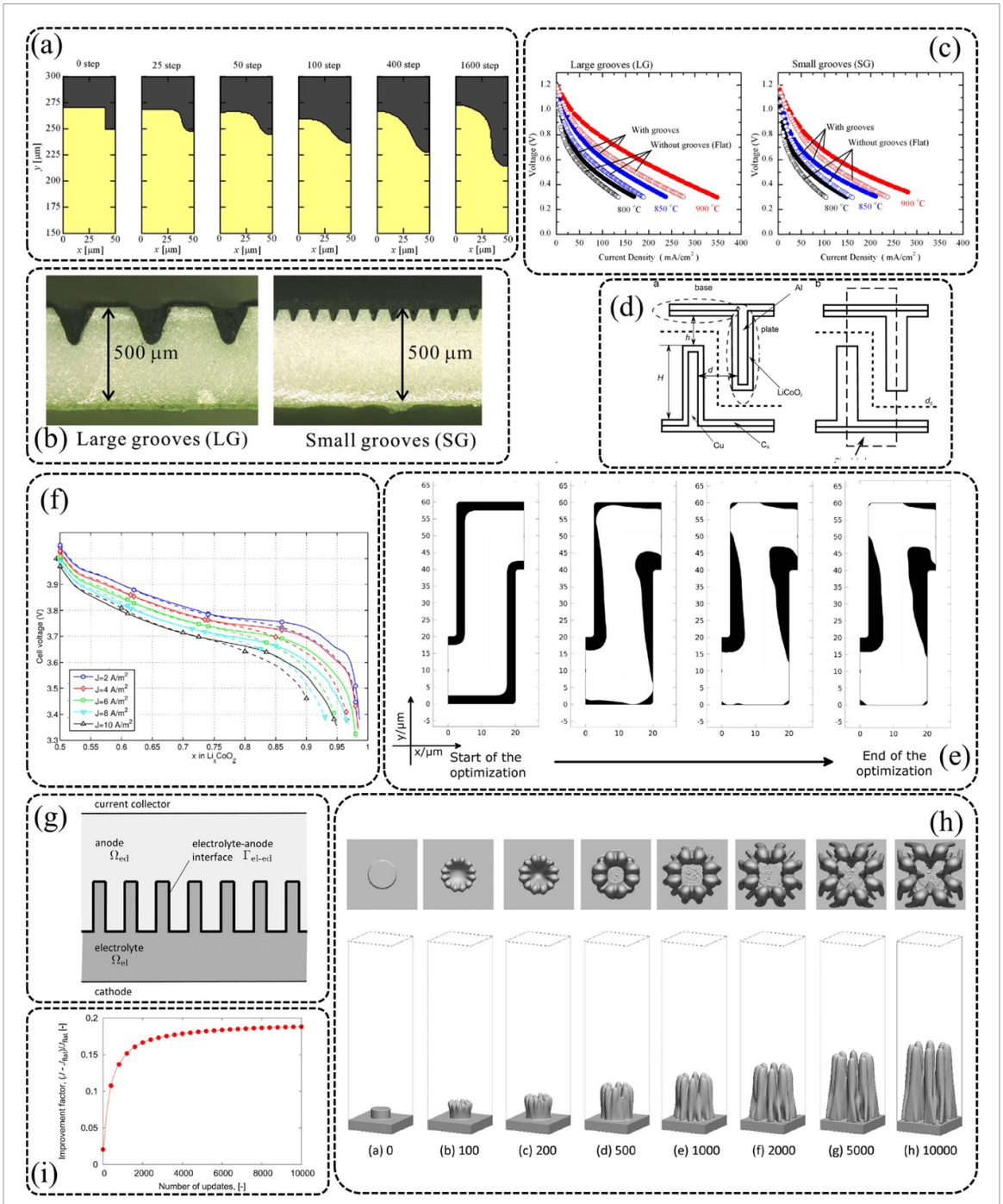

Figure 3. Examples of applying topology optimization based on level-set techniques to optimize the electrode/electrolyte interface in EEDs. Schematics and images show (a) the evolution of the cathode/electrolyte interface of a solid oxide fuel cell during the optimization process, (b) a cross-section view of the cathode/electrolyte interface with small and large grooves, (c) a comparison of IV performance curves of cells with grooved and flat electrolytes (Reprinted from [74], Copyright (2011), with permission from Elsevier); (d) a schematic illustration of a 3D-microbattery considered in the study of applying the level-set method with topology optimization, (e) the evolution of the electrode/electrolyte interface of the 3D-microbattery during the optimization process, (f) discharge curves of cells with optimized and non-optimized electrode/electrolyte interfaces at various current densities, where dashed lines represent the non-optimized interface and solid lines represent the optimized interface (Reprinted from [75], Copyright (2013), with permission from Elsevier); (g) a schematic illustration of the solid oxide fuel cell considered in the study of applying the level-set method with topology optimization representing the anode electrode–electrolyte interface, (h) the evolution of the anode/electrolyte interface of a solid oxide fuel cell during the optimization process, and (i) the convergence history of the optimization (Reproduced from [30]. © 2019 The Electrochemical Society. All rights reserved).

interface. As illustrated in figure 3(b), two configurations with varying groove sizes—small and large—were fabricated, both with an overall thickness of 500 microns. Figure 3(c) compares the experimental I–V curves of modified electrodes with small and large grooves, at various operating temperatures, to that of a conventional electrode. It reveals a noticeable increase in current density across a range of terminal voltages. This finding highlights the practical challenges and opportunities in translating optimized designs into real-world applications. The experiments underscore the potential for performance gains through interface

modification, even when the fabrication does not perfectly align with the theoretical optimization. In 2013, Zadin *et al* [75] embarked on a study aimed at enhancing the design of 3D-microbatteries, as depicted in figures 3(d)–(f). To achieve this, they employed a structural TO based on the level-set method. The investigation centered around optimizing the geometries, displayed in figure 3(d), of the positive electrode ($LiCoO_2$) and negative electrode (LiC_6), separated by a $LiPF_6{\cdot}PEO_{20}$ polyethylene oxide polymer electrolyte. With the idea of obtaining a more uniform electrochemical activity on the electrode surface, the researchers formulated the optimization problem as a function of current density. Moreover, to maintain the volume of electrode material in the cell, a Heaviside function was applied to the level-set variable, helping to control the electrode volume over the optimization course. Following the optimization process, the researchers found that coating the current collectors with active material distributed in a non-uniform manner yielded favorable results (see figure 3(e)). Further analysis compared the performance of the optimized battery designs with those that were not optimized. It was discovered that geometry optimization led to a remarkable increase in cell performance, with improvements of up to 2.25 times observed, as shown in figure 3(f). This significant enhancement was attributed to the mitigation of internal energy losses, which were caused by nonuniformities in the ionic transport occurring within the battery.

Onishi and Shikazono's research group [30, 76–79] stands out among notable research teams that have utilized the level-set TO technique to enhance the performance of SOFCs. Their considered geometries are shown in figure 3(g). Their approach involves considering the spatial distribution of the level-set function as a design variable, resulting in a design space with significant degrees of freedom. To address the challenges associated with this approach, they adopted the adjoint method, wherein adjoint equations are solved to compute the sensitivity of the objective function concerning the design variable. In their initial study, Onishi *et al* [30] discovered that the optimal meso-scale structure for the electrolyte-anode interfaces of SOFCs exhibited multiple branches at the top side and characteristic sub-structures like wrinkles at the bottom side (see figure 3(h)). These wrinkles were found to contribute to performance enhancement by homogenizing the electrochemical potential. Figure 3(i) illustrates that the optimized electrolyte-anode interfaces show an improvement of around 18.8% after 10 000 optimization iterations. Building upon this research, He *et al* [77, 78] employed a similar approach, incorporating local radius constraints, to optimize the cathode porous microstructure of SOFCs made of $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ (LSCF). Subsequently, they extended their efforts [79] to apply TO with multiple level-set methods to the nickel–yttria-stabilized zirconia (Ni-YSZ) anode. Given that a single level-set function can only distinguish between two different phases, the multiphase level-set method was employed to parametrize structures with more than two phases. Simulation results indicated that the optimal microstructure comprised Ni particles embedded into YSZ scaffolds, exhibiting a pillar-like structure along the thickness direction. In addition to the studies focusing on the electrode–electrolyte interface of EEDs, Ishizuka *et al* [80] utilized TO with level-set methods to design anodes placed in an electroplating bath. This application aimed to achieve uniform deposition thickness, a critical factor in ensuring desirable surface qualities in various products. The uniformity of the current density on a cathode was employed as the objective function in this context.

TO employing level-set methods has found widespread applications across various fields, ranging from electroplating to SOFCs and lithium-ion batteries. Typically, level-set methods are employed in scenarios where the focus lies on material interfaces. This preference stems from the fact that the interface between different material phases can be precisely defined by iso-contours of a level-set function. This implicit function provides a clear delineation of boundaries, facilitating accurate representation of interfaces. Depending on how the interface is represented in the physical model, using level-set methods can enhance the accuracy of mechanical response predictions near boundaries. Additionally, employing level-set methods helps to mitigate the uncertainties associated with intermediate material phases, a challenge often encountered when utilizing density-based approaches. This inherent capability of level-set methods contributes to their widespread adoption in TO tasks.

3.2. Density-based methods

Building on a previous work [74], two years later, Song *et al* [81] initiated a different modeling and optimization perspective. Iwai *et al* [74] treated the cathode-gas as a homogeneous porous medium and aimed to find the optimal shape for the electrode–electrolyte interface. On the other hand, the subsequent research by Song *et al* [81] shifted focus to the cathode-gas interface itself. The authors limited the scope of their study to a specific electrode configuration consisting of a mixed ionic–electronic conducting material deposited by infiltration onto an ionically conducting scaffold. In such a case, their model predicted that a larger perimeter and a greater amount of scaffold material, regardless of the specific electrode structure, resulted in lower Ohmic resistance. Consequently, to isolate the effect of shape exclusively during the optimization process, the authors introduced isoperimetric constraints on both the perimeter and the amount of material used. The researchers utilized a formulation of TO based on the SIMP method. Their

approach involved design-dependent boundary conditions, necessitating a dynamic treatment of material boundaries within the optimization process rather than predefined delineations. Consequently, specialized methodologies were employed to address the implications of design-dependent boundary conditions. The investigation revealed notable enhancements in performance ranging from 18% to 50% across varied geometrical dimensions and material properties compared to conventional column designs. This underscores the considerable potential for performance improvement through meticulous organization of the cathode microstructure, yielding intricate configurations that were not realizable without a robust mathematical scheme. In addition to the efforts initiated by other researchers, Mathieu-Potvin and Gosselin [82] employed density-based methods to optimize platinum distribution in polymer electrolyte fuel cells (PEFCs). Their primary aim was to maximize current density while maintaining a fixed total amount of platinum. The findings of their study unveiled that the most effective design showcased a gradient-based distribution, concentrating the majority of platinum near the membrane layer. It is crucial to acknowledge that Mathieu-Potvin and Gosselin simplified the system by assuming high conductivity in the solid phase, enabling rapid electron transport and eliminating Ohmic losses related to electron transport. Furthermore, their design domain mesh consisted of only 50 nodes along the in-plane direction and 15 nodes along the through-plane direction, indicating a relatively coarse discretization. A key limitation acknowledged by the authors is their model's inability to account for liquid water transport. This omission leaves the significant negative impact of flooding, caused by concentrating catalyst material in a confined space near the membrane, unaddressed in the results. Despite these simplifications, their research shed light on the potential advantages of optimizing platinum distribution in PEMFCs for enhanced performance. Meanwhile, Lamb *et al* [83–87] tackled a similar topic by optimizing catalyst distribution in PEFCs with the computational domain depicted in figure 4(a). In these studies, TO is performed by allowing the catalyst amount to vary independently at each location within the design domain. This freedom translates to a significant increase in the number of design variables compared to Mathieu-Potvin and Gosselin [82], exceeding 10^4, since the catalyst amount at each node of the finite element mesh becomes an optimization variable. They also considered the impact of land and channel on optimized catalyst distribution. In their findings, Lamb *et al* recommended placing more catalyst material (i.e. platinum) under the gas channel than under landings and toward the membrane interface, as seen in figure 4(b), might improve the output power. Figure 4(b) also compares the optimized catalyst distribution under two different overall loadings of 0.1 and 0.2 mg cm^{-2}. While the exact distributions are different, both cases show a similar increasing incremental trend when moving from the gas diffusion interface toward the membrane side. However, it is important to note that they solely considered the amount of catalyst as a design variable, without considering the reorganization of porosity and ionomer distributions, which could potentially benefit cell performance. Evidently, since the total volume fractions of all materials in an electrode must add up to unity, optimizing the amount of one component requires flexibility in another. For instance, if the amount of ionomer is fixed and uniform throughout the electrode, then the porosity needs to be freely adjustable when optimizing the catalyst loading. Lamb and colleagues acknowledged the limitation of their study in simultaneous optimization of multiple materials and successfully addressed it in 2020. In their recent studies [86, 87], they tackled the challenge of optimizing the distribution of multiple components within the catalyst layer, including platinum particles (catalyst material), Nafion polymer (ionic conductive material), carbon (catalyst support and electric conductive material), and porosity. They achieved this by treating the volume fraction of each constituent as a design variable at every location within the layer. However, managing such a complex optimization problem presented a significant computational hurdle. To address this, the authors employed an adjoint variable method. Particularly [87], involved optimizing the distribution of all constituents (catalyst, Nafion, carbon) simultaneously. Similar to the individual optimization of various constituents that was presented in [86], porosity was not directly optimized. The results consistently demonstrated that a higher volume fraction of catalyst and electrolyte material near the membrane was favorable as the proxy to the ion exchange membrane improves the overall electrochemical reaction rate. Additionally, higher porosity at the opposite side of the catalyst layer (near the diffusion layer) and under the channel area enhances oxygen delivery, ultimately boosting overall performance. It is important to note that, similar to the work by Chen [36], their model does not account for two-phase flow, neglecting the transport of liquid water within the catalyst layer. In addition, since the models used in these studies are 2D, the impact of longitudinal direction on the final optimum design has not been well studied.

In addition to the advancements made in PEFCs, Mitchell and Ortiz [88] ventured into applying density-based TO to design optimal multifunctional silicon anode structures for lithium-ion batteries, aiming to develop next-generation high-performance secondary batteries. While the silicon anode holds promise due to its inherent high capacity for storing lithium ions, its structures undergo a substantial 310% volume expansion upon lithiation, leading to severe damage such as active particle pulverization and disconnected charge transport paths. Furthermore, the low intrinsic electric conductivity of silicon results in

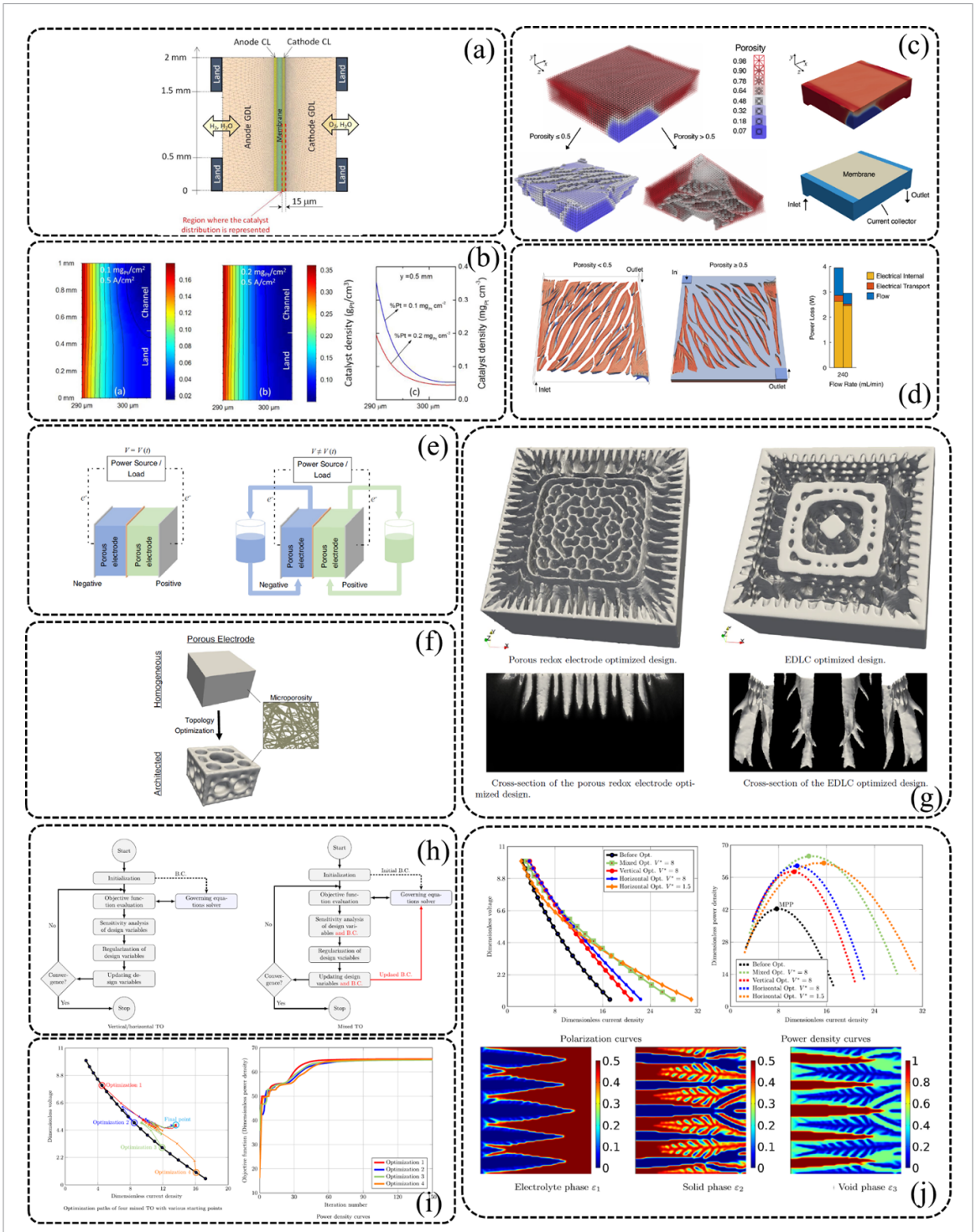

Figure 4. Examples of applying topology optimization based on density-based methods to optimize the electrode in EEDs. Schematics and images show (a) the computational domain of a proton exchange membrane fuel cell considered in the study of applying the density-based method with topology optimization, (b) optimized catalyst distributions for two different Pt loadings and the 1D optimum platinum distribution at the middle of the computational domain (Reproduced from [83]. © The Author(s) 2017. Published by ECS. CC BY 4.0.); (c) the computational domain of a negative half-cell compartment of a vanadium flow battery by which the electrode is split into portions with a unit cell, (d) optimized porosity distribution of the electrode (Reproduced from [92], Copyright (2021), with permission from Elsevier); (e) two systems considered in the study of, which are a device operates through redox reaction and a device operates through charge storage using a double layer, (f) schematic diagram showing the porous electrode before and after optimization in which the material seen is, in fact, porous, (g) 3D optimized designs of two systems considered in the study (Reproduced from [31], Copyright (2022), with permission from Springer Nature); (h) flowchart of the algorithm for conventional and mixed topology optimization, (i) convergence history of mixed topology optimization with various starting points showing its self-guidance feature, and (j) performance curves as well as the optimal volume fraction distribution of each material constituent (Reproduced from [32], Copyright (2023), with permission from Elsevier).

poor rate performance due to sluggish electron transport through the material. To tackle these structural and charge conduction design challenges, Mitchell and Ortiz employed TO methods. Initially, they considered the objectives individually and later extended the methodology to a bi-objective formulation to simultaneously address both the structural and conduction design criteria. Through their research, they discovered that a rigid frame structure served as an excellent compromise between the structural and conduction design criteria, offering both the required structural rigidity and direct conduction pathways. In a recent work by Pejman and Raeisi Najafi [89], the authors introduced a novel approach for multi-objective TO aimed at crafting Structural Battery Electrolytes (SBE) within multifunctional structural battery composites. The study aimed to overcome the inherent conflict in achieving both high mechanical strength (stiffness) and high ionic conductivity, while also minimizing heat generation in the electrolyte, which are crucial factors for maximizing battery performance. The researchers implemented a multiphysics, multi-objective gradient-based approach to simultaneously maximize both the ionic conductivity and stiffness of the SBE. To ensure prevention of overheating, the optimization process incorporated constraints on the maximum allowable temperature and void volume fraction. The proposed design framework integrates electrochemical, thermal, and structural physics, enabling the creation of an optimized SBE microstructure. The study investigated two optimization scenarios, including (1) bulk condition, and (2) carbon fiber included condition. The former focused on optimizing the microstructure of SBE assuming a bulk material, where carbon fibers were not explicitly included as part of the design solution. However, the latter scenario built upon the first by introducing carbon fibers as an explicit element within the design space. It is important to note that the carbon fibers were treated as fixed, non-optimizable elements during the optimization process. In both cases, two materials, including a solid phase and a compliant phase, were used. While the solid phase ensured high stiffness and thermal conductivity, the compliant phase enabled efficient electric charge transport. The study successfully generated a set of Pareto optimal microstructures with various trade-offs between effective ionic conductivity and compliance. Additionally, explicit incorporation of carbon fibers within the design space significantly altered the optimized SBE design compared to the bulk scenario.

Meanwhile, thermal mismatch significantly influences the stress state and lifetime of SOFCs. In response, Li *et al* [90, 91] endeavored to mitigate this issue by designing LSM–YSZ cathodes and Ni–8YSZ anodes using density-based TO. The microstructures of the cathode and anode took the form of periodic fiber bundles. The results demonstrated that the coefficients of thermal expansion of these microstructures closely matched those of the electrolyte layer at different temperatures, effectively eliminating thermal mismatch issues.

Inspired by the configuration of electrodes in redox flow batteries (RFBs), where the electrode typically comprises a disordered, homogeneous assembly of micron-scale electroactive particles like carbon fibers and felts, Beck and Worsley's research group [31, 92] stands out as a prominent contributor in applying TO to enhance porous electrode structures. The primary goal behind their design philosophy is to maximize surface reactions while minimizing overpotential and hydraulic losses. In their initial study, Beck *et al* [92] focused on restructuring porosity distributions with the objective of minimizing power loss and creating electrodes with engineered porosity distribution, as shown in figure 4(c). A comparison between these architectured electrodes (demonstrated in figure 4(d)) and bulk, uniform porosity electrodes revealed enhanced power efficiency across various flow rates and currents. Continuing their investigation, Roy *et al* [31] illustrated a framework of TO in designing porous electrodes for two applications: one involving a porous electrode driving a steady Faradaic reaction (as seen in RFBs), and the other operating transiently without a Faradaic reaction (like in electric double layer capacitors), as depicted in figure 4(e). In their research, they utilized a porous model (illustrated in figure 4(f)), wherein the porous material is termed microporous to differentiate it from the larger pores formed through the optimization process. Across all scenarios, the optimized designs exhibited superior performance compared to undesigned, monolithic single porosity electrodes. In the case of RFBs, this translated to overpotentials reduced by up to 84%, while the electric double layer capacitor electrode showed energy losses reduced by up to 98%. Moreover, they demonstrated the versatility of these techniques by extending them to a three-dimensional electrode design (displayed in figure 4(g)), paving the way for manufacturing and testing high-performance architected electrodes. This exploration holds significant promise in advancing the efficiency and functionality of porous electrode systems across various electrochemical applications. However, the model developed by Roy *et al* [31] has limitations regarding its consideration of concentration effects and hydraulic requirements. In systems with convective-reactive transport, like those in RFBs, the concentration of the solution flowing through the electrodes depends on the flow rate. This flow rate in turn affects the pressure drop needed to pump the solution (hydraulic requirements). Higher pressure drop requirements mean more power loss by the cell-pump system, which could affect the net generated power and should be considered in the optimization objective function. Another key factor is the difference between the concentration of the solution in the bulk and at the surface of the electrodes. Electrochemical reactions happen at the electrode surface, so for accurate simulations, it is necessary to distinct surface and bulk concentrations. This difference can be accounted for by including a

mass transfer coefficient, which typically depends on the flow velocity. However, the authors oversimplified the modeling and optimization by ignoring the concentration effect and hydraulic requirements.

In a recent study, Charoen-amornkitt *et al* [93] explored the application of TO in the design of anode catalyst layers for proton exchange membrane water electrolyzers (PEMWEs). They focused on a 2D electrochemical porous electrode model, which encompasses various processes such as water transport, species diffusion, electric charge transport, ionic charge transport, and redox reactions. The porous electrode was conceptualized to consist of three phases: a void phase, an electrolyte phase, and a carbon-supported catalyst phase. Their optimization approach aimed to maximize cell performance by adjusting the volume fractions of the electrolyte and carbon-supported catalyst materials. The results revealed that introducing a heterogeneously-distributed structure led to more efficient cells. During the optimization process, changes in the volume fraction of constituent materials occurred, resulting in the formation of a zigzag interface in the reactor. This interface facilitated a sufficient supply of charges required for the electrochemical reaction. Compared to cases with uniform spatial distribution of constituent materials, the proposed heterogeneous structure demonstrated a notable enhancement in the electrochemical reaction, achieving approximately a 40% improvement. This highlights the effectiveness of TO in enhancing the performance of PEMWEs by optimizing the electrode design. Despite this, the authors simplified the system by disregarding the effects of oxygen bubble formation within the electrode, among other factors, resulting in a uniform distribution of porosity that eliminates the necessity for an oxygen bubble removal pathway. Additionally, their research indicates that distinguishing between electronically conductive materials and the catalyst, coupled with utilizing multi-objective optimization to minimize the catalyst volume fraction, has the potential to significantly reduce catalyst usage. While fuel cells have a longer history of modeling, water electrolyzers are catching up. Researchers are adapting their knowledge from fuel cells to improve existing electrolyzer models. However, water electrolyzers involve additional complexities, like bubble formation [94], which need to be factored in. The coverage of reaction sites by these bubbles is an additional limiting factor that requires detailed consideration of the bubble evolution process, such as nucleation and bubble growth. This means there is a need not only for better optimization methods, but also for more advanced modeling techniques to create robust and reliable electrode designs for electrolyzers. To overcome some of the limitations mentioned earlier, Passakornjaras *et al* [95] optimized the anode catalyst layer of a PEMWE, accounting for limitations related to gas coverage and effects of temperature distribution. Although their modeling of physical phenomena differs, the optimization process is similar to that in [93]. The resulting optimized structures significantly outperformed a homogeneous electrode design, with electrode current densities 2.7 times higher at high operating voltage (2.03 V) and 1.2 times higher at low operating voltage (1.73 V). Future research on TO for PEMWEs should incorporate more advanced models of the various physical and chemical phenomena occurring in the electrode and consider the durability of optimized structures under thermal and mechanical stress. Additionally, exploring multi-objective optimizations to balance reaction rates and material usage may be necessary.

While TO has been widely used to search for optimized electrode structures in EEDs, previous studies have not achieved maximum output power for power sources. This limitation arises because researchers are typically constrained to either minimizing overpotential at a fixed current density or maximizing current density at a specified overpotential. While both strategies can improve performance in terms of power density, they do not necessarily lead to maximum output power. Alizadeh *et al* [32] proposed a novel mixed TO approach to enhance the performance of these systems by simultaneously modifying the electrode structure and the working conditions (see figure 4(h)). Unlike conventional approaches, this method focuses on enhancing the maximum power point. Their research demonstrates that the mixed TO approach outperforms conventional vertical and horizontal optimizations, where either terminal voltage or current density is optimized. Additionally, it has been tested under various starting points, as shown in figure 4(i), consistently yielding the same output. This self-guidance feature of this method eliminates the need for a prior decision on the optimization starting point. Figure 4(j) reveals that the optimal distribution of materials within the design domain resembles a complex tree-root-like structure. The formation of diffusion channels, facilitated by high concentrations of voids in certain parts of the system, enables the delivery of reactant material across the entire system. This structural pattern bears resemblance to the vascular layout observed in nature, such as in the leaves of plants. The network of veins in plant leaves, which transports water and nutrients, exhibits a similar structural pattern.

Charoen-amornkitt *et al* [33] utilized TO to engineer porosity distributions within a system characterized by a 1D nature. In this system, the concentration remains constant at $x = 0$, and there are no fluxes at $x = L$. By exploring various dimensional models, they discovered that increasing the design dimensionality beyond one enhances system performance by minimizing entropy generation. However, they observed a relatively modest performance improvement when transitioning from 2D to 3D designs. This led to the hypothesis that due to the inherently 1D nature of the problem, a 2D model suffices to significantly enhance performance.

Building on this insight, Long *et al* [96] focused on systems with a 2D flow nature, specifically investigating the impact of rib structures and electrode thickness. They evaluated three different geometries, including one with fixed species concentration at the inlet and zero-flux boundary conditions, effectively confining the system to a 1D nature. To increase the nature of the species diffusion to a 2D flow characteristics, they introduced a rib at the inlet. Beneath the rib, a region restricted reactant movement while allowing electron transfer. Additionally, they reduced the thickness to encourage more 2D flow characteristics. Their findings indicated that in rib-containing cases, reduced inlet area limited species availability, a challenge mitigated by decreasing reactor thickness. Examining the optimized porosity distribution, they observed the formation of diagonal channels in systems exhibiting 2D characteristics. Interestingly, augmenting the system model's dimensionality beyond its inherent nature did not significantly impact the reaction rate.

Until now, with a few exceptions, prior research primarily focused on maximizing the reaction rate within the designated design domain. However, as previously discussed, there exists an alternative approach: minimizing overpotential at a fixed current density. Alizadeh *et al* [97] undertook a comparative analysis of two optimization strategies aimed at enhancing the performance of electrochemical reaction–diffusion systems, as displayed in figures 5(a)–(c). These strategies involved minimizing overpotential at a fixed current density and maximizing current density at a specified overpotential. The researchers analyzed a 2D triple-material electrode, similar to the catalyst layer found in PEFCs. However, their model simplified the processes occurring in PEFCs by neglecting the two-phase flow of gas and liquid water, a common phenomenon in low-temperature fuel cells. The electrode consisted of solid, electrolyte, and void phases. While the solid phase was responsible for electron transport and provided necessary reactive surface area, the two other phases facilitated ion transport and mass diffusion. The optimization aimed to find the best distribution of constituents volume fractions through a density-based method. The resulting optimal layouts exhibited intricate root-like structures (see figure 5(a)), which facilitated transport processes and led to a remarkable improvement in the conversion rate of up to 116.7%. Further analysis revealed that the optimal layout varied significantly depending on the dominant processes at different voltages (or current densities). For instance, when the optimization focused on low voltages (high current density), where concentration overpotential limits performance, the algorithm favored designs with higher porosity and larger diffusion channels. Conversely, optimization at high voltages (lower current density) resulted in a design with a higher solid phase volume fraction distributed throughout the electrode. This increased the reactive surface area, thereby reducing activation overpotential.

To the best of our knowledge, density-based methods are more prevalent than level-set methods in optimizing the structure of electrodes for EEDs, as evidenced by the abundance of studies employing this approach. A closer examination of these studies reveals a fundamental difference in the focal point of interest compared to those utilizing level-set methods. Density-based methods primarily concentrate on optimizing the distribution of materials within the electrode structure rather than delineating the interface between different materials. These density-based methods have found applications across a wide range of EEDs, spanning from power sources like PEFCs and SOFCs to power-consuming devices such as PEMWEs, as well as energy storage systems like lithium-ion batteries and RFBs. However, it is important to note that the treatment of material density may vary across different studies employing density-based methods. In density-based methods, material density typically ranges from 0 to 1, signifying regions containing a mixture of material and void. While this 'gray area' typically holds no physical meaning in structural mechanics applications, it holds physical significance in the context of EEDs, particularly in porous models used at the macroscale level. Here, material density values between 0 and 1 are meaningful and directly inform the modeling process. To address the gray area and ensure meaningful material distribution for structural mechanics problems, projection methods are often employed. However, in EEDs, where porous models are prevalent, material density values between 0 and 1 are utilized and hold relevance in representing the actual physical structure of the electrode materials by assigning them as macroscopic properties, like volume fraction and porosity.

3.3. Connection between entropy generation minimization and TO

TO plays a pivotal role as a mathematical tool in the intricate design and optimization of complex structures. However, it is crucial to acknowledge the numerical nature of the solutions derived from TO, which are influenced by various factors including the choice of objective function, algorithmic approach, and tuning parameters. Consequently, it becomes imperative to establish a robust theoretical framework to underpin these optimized solutions. In addressing this need, Tsushima's research group [32, 33, 93, 96–99] advocates for the integration of principles derived from entropy generation minimization theory. Inspired by the widespread use of entropy generation analysis in evaluating thermal systems, their objective is to align with a system characterized by minimal entropy generation. This approach aims to lay the groundwork for a design methodology that is firmly grounded in physical principles, thereby reducing reliance on arbitrary tuning

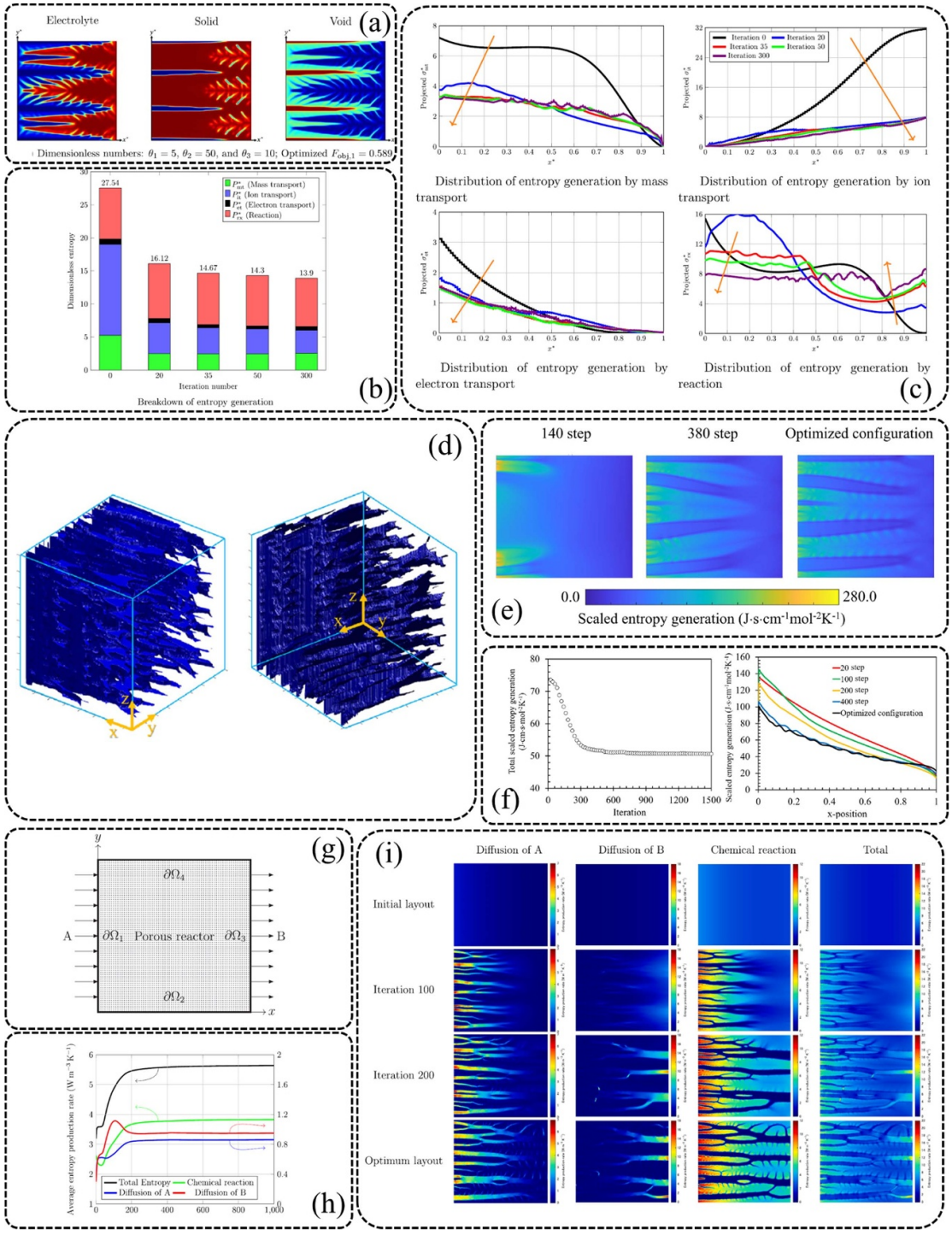

Figure 5. Examples of works attempting to draw connections between topology optimization and entropy generation minimization. Schematics and images show (a) optimized volume fraction distribution of material constituents in electrochemical porous reactors, (b) global entropy generation of each phenomenon occurring in porous reactors, approaching minima post-optimization, (c) 1D projected entropy generation distribution of each phenomenon in porous reactors, displaying increased uniformity akin to the equipartition principle (Reproduced from [97].© 2023 The Author(s). Published on behalf of The Electrochemical Society by IOP Publishing Limited. CC BY 4.0.; (d) 3D optimized porosity distribution from different angles, where surfaces represent areas with porosity higher than 0.95, (e) scaled entropy generation distribution during optimization, (f) history of global scaled entropy generation and 1D projected scaled entropy generation distribution, showcasing the approach towards minima and a more uniform distribution resembling the equipartition principle (Reprinted from [33], Copyright (2023), with permission from Elsevier); (g) computational domain of two-species reaction–diffusion system, (h) history of global entropy generation during the optimization process, and (i) spatial distribution of entropy generation by various mechanisms at different iterations (Reproduced from [98], Copyright (2023), with permission from Elsevier).

parameters that are usually necessary for optimization algorithms (e.g. filter radius). Entropy generation analysis offers a rigorous framework for assessing the thermodynamic efficiency of systems, enabling the identification of optimal designs that minimize energy wastage and maximize performance. By

incorporating these principles into the design process, it is anticipated that the resulting electrode structures will exhibit heightened reliability and effectiveness. This is because they are founded on fundamental physical principles rather than ad hoc parameter adjustments.

Charoen-amornkitt *et al* [33] embarked on a comprehensive research endeavor, commencing with the utilization of TO to optimize the structure of porous electrodes (see figures 5(d)–(f)). To evaluate the entropy generation during this optimization process, they employed entropy generation analysis, recognizing the complexity of assessing entropy generation in the porous media of EEDs. The nonequilibrium nature of these systems, compounded by the presence of chemical reactions, posed significant challenges. In response, they adopted nonequilibrium thermodynamics as a theoretical framework, enabling the assessment of local entropy generation rates in systems not in a state of global equilibrium. In their study, the researchers simplified the system by focusing solely on a porous reactor within reaction–diffusion systems involving a single species. As the optimization progressed with the objective of maximizing reaction within the design domain, entropy generation inevitably increased. To facilitate comparison, they introduced scaled entropy generation, mitigating the influence of the increasing objective function. An essential aspect of their investigation was to examine the effects of design dimensionality on optimization. They found that while 0D and 1D optimization results exhibited little difference in overall reaction, significant increases in reaction were observed when 2D or 3D optimization was permitted. This phenomenon stemmed from the emergence of a geometrically intricate diffusion field reminiscent of biological structures in 2D or 3D optimization. The 3D optimized porosity distribution obtained from their work is illustrated in figure 5(d). As the optimization process advanced, not only did the global scaled entropy generation approach a minimum, but the distribution of scaled entropy generation in 1D also gradually transitioned towards greater uniformity (see figures 5(e) and (f)). These findings align with the equipartition principle, indicating that uniform entropy generation across space may result in less dissipation, leading to the minimum entropy generation rate and a thermodynamically optimal design.

In the subsequent phase of their research, Alizadeh *et al* [98] expanded their investigation to encompass a reaction–diffusion system involving two species operating in a steady-state mode, as depicted in figure 5(g). They utilized a density-based TO algorithm, which resulted in a remarkable 57% improvement in system performance compared to a uniform layout with equivalent average porosity. Throughout the optimization process, the researchers closely monitored the porosity distribution, concentrations of reactant and product substances, and reaction rates. The study's outcomes underscore the importance of achieving a delicate balance between diffusion and reaction mechanisms to enhance performance. This equilibrium was achieved through the formation of primary and secondary channels within the reactor during the optimization process. The optimized porosity distribution, derived from the algorithm, exhibited a tree-root-like configuration, resembling patterns observed in previous research [33]. Similar to their earlier study, this research delved into dissecting the contributions to system entropy generation and quantifying them throughout the optimization procedure (see figures 5(h) and (i)). The findings revealed that the optimized design solution corresponded to the minimum scaled entropy generation resulting from chemical reactions. The researchers suggested that these findings could have significant implications for advancing the understanding of the theoretical upper limit of reaction–diffusion system performance, regardless of the inherent limitations of optimization methods.

Additionally, Alizadeh *et al* [97] also introduced an entropy generation model to quantitatively evaluate irreversibilities in a system involving transport phenomena and electrochemical reactions. By analyzing entropy generation trends for both optimization approaches, the research provided insights into optimizing the distribution of constituents in porous electrochemical reactors and elucidated the relationship between TO and entropy generation rate. As depicted in figures 5(b) and (c), the findings aligned with principles of entropy generation minimization [100] and equipartition of entropy production [101, 102]. It is noteworthy that when employing the strategy of maximizing current density at a specified overpotential, scaling of entropy generation is necessary to counteract the escalating entropy flux during optimization. However, the proper scaling becomes more complicated with additional physics introduced to the system. In contrast, minimizing overpotential at a fixed current density corresponds to reducing entropy generation without requiring scaling. This distinction arises intuitively as overpotential is commonly associated with irreversible losses within EEDs.

In various applications, researchers have observed intricately distributed pores that resemble tree-root-like structures, reminiscent of natural patterns such as root systems in trees and respiratory networks in lungs. These natural systems are characterized by efficient mass transport mechanisms, suggesting that exploring similar structures in engineered systems could enhance our understanding of natural design principles. The studies discussed above utilize TO techniques to design electrodes for electrochemical energy storage and conversion systems. By integrating these optimized structures with established theories like entropy generation minimization, researchers aim to uncover connections between

engineered designs and natural phenomena. With a design reminiscent of nature, it may very well be the optimal design we are in search of, acknowledging that nature tends to design its systems in the most effective manner. By drawing parallels between topologically optimized structures and entropy generation minimization, researchers seek to unravel the underlying principles governing natural design. This area of research has the potential to bypass the optimization process by minimizing the need for repetitive evaluation of the objective function and governing equations. By doing so, it could significantly reduce the computational resources and the reliance on tuning parameters typically required in optimization tasks. This interdisciplinary field has the potential to revolutionize the design of various technologies by harnessing insights from nature's efficient solutions. Ultimately, by emulating nature's design strategies, humanity may unlock new avenues for innovation and enhance the performance of engineered systems.

One of the significant contributions in this field was made by Long *et al* [103]. In their study, Long *et al* [103] derived exact solutions for 0D optimization of reaction–diffusion systems, focusing on both maximization (maximizing the reaction) and minimization (minimizing the concentration at the boundary) problems. Their work provided critical insights into the relationship between optimized structure and entropy generation. Following this, they extended their research to 2D and 3D TO to explore the characteristics that an optimized system should possess. They projected the concentration distribution into a 1D representation, identifying that the key characteristic of an optimized system is a linear concentration distribution. Based on this observation, they developed a design theory suggesting that an optimized system should exhibit a linear concentration distribution. By assuming that the optimized structure consistently produces a linear concentration distribution, they were able to substitute this assumption into the governing equations to directly solve for the porosity distribution. This approach allowed them to obtain the porosity distribution without relying on an iterative optimization process. However, it is important to note that this assumption was primarily applied to 1D optimization. Further research is necessary to extend these findings to 2D and 3D optimization, as the complexities introduced in higher dimensions may require additional considerations and refinements to the design theory.

4. Other notable numerical studies in electrode structural optimization

While the primary focus of this paper is formal implementation of TO methods, as described in section 2, for innovative electrode design in EEDs, other approaches exist for optimizing electrode morphology. Over the years, researchers have utilized various mathematical techniques, such as parametric optimization and functionally graded methods, to improve the topographical design of electrode structures. Parametric optimization aims to find the optimal value(s) for one or more macroscopic properties (e.g. porosity) of a homogenous design to enhance overall cell performance. While TO adjusts material distribution locally, resulting in a heterogeneous design, parametric optimization operates at a global level, fine-tuning design variables. Examples include optimizing Nafion loading in PEFC catalyst layers for maximized power output or finding the ideal porosity or fiber diameter in RFB electrodes to balance reactive surface area with hydraulic permeability. While simpler to implement, these methods are limited in generating highly efficient designs. Functionally graded methods are more advanced approaches building upon parametric optimization. It can involve: (1) dividing the electrode design domain into multiple regions and performing independent parametric optimization on each [104], and (2) utilizing pre-defined distributions based on mathematical functions (e.g. sinusoidal or polynomial) [105]. Despite offering greater design freedom than standard parametric optimization, functionally graded methods are still limited by the number of pre-defined domains or functionalities. In contrast, TO provides a robust framework for automatically generating material distributions with high resolution. It is noteworthy that approaches like functionally graded design differ from the formal TO which automatically manipulates the material distribution itself. Nonetheless, the algorithmic approaches share similarities, as they both offer a heterogeneous material property distribution for improved performance. While this review focuses on TO, we briefly mention some noteworthy works that have employed mathematical techniques for morphological modification and improved electrode performance. These studies offer valuable insights for interested readers but are not in any way inclusive.

He *et al* [106], for instance, investigated the effect of catalyst layer design on the performance of a PEFC. The researchers developed a macroscopic 3D multiphase non-isothermal model of PEFC to simulate the cell behavior under various conditions. The study aimed to understand how different design parameters, such as platinum (Pt) loading, platinum-to-carbon (Pt/C) ratio, ionomr-to-carbon (I/C) ratio, carbon particle radius, and electrochemical specific area (ECSA) of platinum particles, influence oxygen transport resistance, performance, water transport, and the oxygen transport process within the electrode. Through their numerical simulations and analysis, the researchers found significant correlations between the electrode design parameters and the performance of the cell. They observed that variations in Pt loading, Pt/C ratio,

and I/C ratio directly impacted the oxygen transport resistance within the catalyst layer. Additionally, they highlighted the importance of considering the carbon particle radius and ECSA in optimizing the performance of PEFCs. While the study is primarily a parametric sweep analysis, rather than an algorithmic optimization, it provided valuable insights into the complex interplay between design parameters and performance metrics in PEFCs, offering a foundation for further research and development in the field of fuel cell technology. In a different application, Tsushima and Suzuki [107] used a bound optimization by quadratic approximation algorithm to simultaneously optimize fibrous electrode architecture of a vanadium RFB. The multi-parameter optimization included porosity, fiber diameter, and electrode thickness as well as two other channel-related geometrical parameters. Each of these parameters were allowed to vary in a given range. Their results showed that a combination of thinner fibers and thicker electrodes could enhance the overall cell performance thanks to a higher reactive surface area. However, this should be accompanied with a relatively high porosity ($\sim$0.89) to facilitate electrolyte flow that can directly affect the active species transport resistance between bulk solution and solid/liquid interface. The authors suggested simultaneous optimization of various electrode parameters are crucial for a comprehensive design with boosted performance. Functionally graded electrode design has been used for various applications, such as fuel cells and batteries [104, 108]. Srinivasarao *et al* [109] used a multi-layer design of catalyst layer to maximize the generated current density and minimize platinum loading. To achieve this, the researchers considered an innovative design with four catalyst layers as shown in figure 6(a) and optimized various design variables such as the platinum loading, ionomer loading, weight fraction of platinum on carbon, and thickness of layers under a range of cell voltages from 0.4 to 0.7 V. By optimizing these parameters for each layer, the study sought to achieve cost reduction and performance enhancement in comparison to the base case design with uniform material distribution. The findings revealed that the PEFC with multiple catalyst layers exhibited superior performance compared to an optimized PEFC with a single catalyst layer across all operating voltages. Under all operating conditions, the optimization favored a higher porosity in the layers closer to the gas diffusion layer (see figure 6(b)). The optimal ionomer volume fraction, on the other hand, shows an opposite trend with more ionomers concentrated in the layer neighboring the membrane (see figure 6(c)). Additionally, the study demonstrated a significant reduction of 17% to 60% in platinum loading with an increase in the number of catalyst layers for low and high current density regions, respectively.

Although the formal implementation of TO typically involves coupling an optimization algorithm with a continuum macroscopic model to find the best distribution of continuous variables, applying topological optimization to electrode structures extends beyond this method. Various phenomena at micro- or nano-scales take place at electrodes of EEDs. Geometrically resolved models can describe these intertwined multiphysics phenomena at a higher resolution. However, the high computational cost of approaches like direct numerical simulation (DNS) of geometrically resolved structures has posed challenges in integrating these models with TO algorithms. As computer technology advances and more efficient modeling and mathematical schemes are developed, a new trend is emerging. Pore- and particle-scale models are now combined with optimization algorithms for morphological optimization of porous electrodes. An exemplary instance is the use of TO with the LBM. Previously applied in thermofluidic systems [111–114], TO using LBM has recently been extended to electrode design for RFBs [110]. Tanaka *et al* [110] focused on fluid behavior in fibrous porous electrodes and optimized electrode structure using adjoint-state LBM. By updating solid and fluid distribution through a level-set function, the optimization algorithm aimed to minimize concentration flow rate at the outlet of a domain with a resolution of 1 μm per voxel. Figure 6(d) compares the structure and vanadium ion concentration distribution of an ordered fibrous electrode with those of optimized one. According to figure 6(e), compared to an ordered structure with fiber diameter of 4 microns, the optimized structure had a lower cost function value by about an order of magnitude in only 25 iterations. The optimization boosted reaction rate by minimizing the amount of active species leaving the electrode before reacting, leading to a more efficient process. Another research direction for designing electrodes with engineered microstructure, initiated by Forner-Cuenca's research group [34, 115], employs pore network modeling (PNM) together with metaheuristic optimization algorithms. This approach seeks optimal pore network topology to enhance cell performance. Despite methodological differences with level-set or density-based TO, they also investigate how a heterogeneous design of porous electrodes could benefit overall cell performance. To achieve higher resolution not attainable through macroscale models and to mitigate the intensive computational cost of DNS methods, authors initially developed a PNM model of RFB (see figure 6(f)). With the goal of building a predictive design framework, the developed PNM model is coupled with a genetic algorithm to optimize network morphology based on a bottom–up design approach. In their first attempt [34], they employed a cubic lattice as shown in figure 6(f) and manipulated the pore and throat size distributions without changing the pores positions. The optimized structures exhibit improved fluid distribution through the formation of a bimodal pore size distribution (see pore size distribution in figure 6(f)), resulting in preferential longitudinal flow pathways (see figure 6(g)) and a 73%

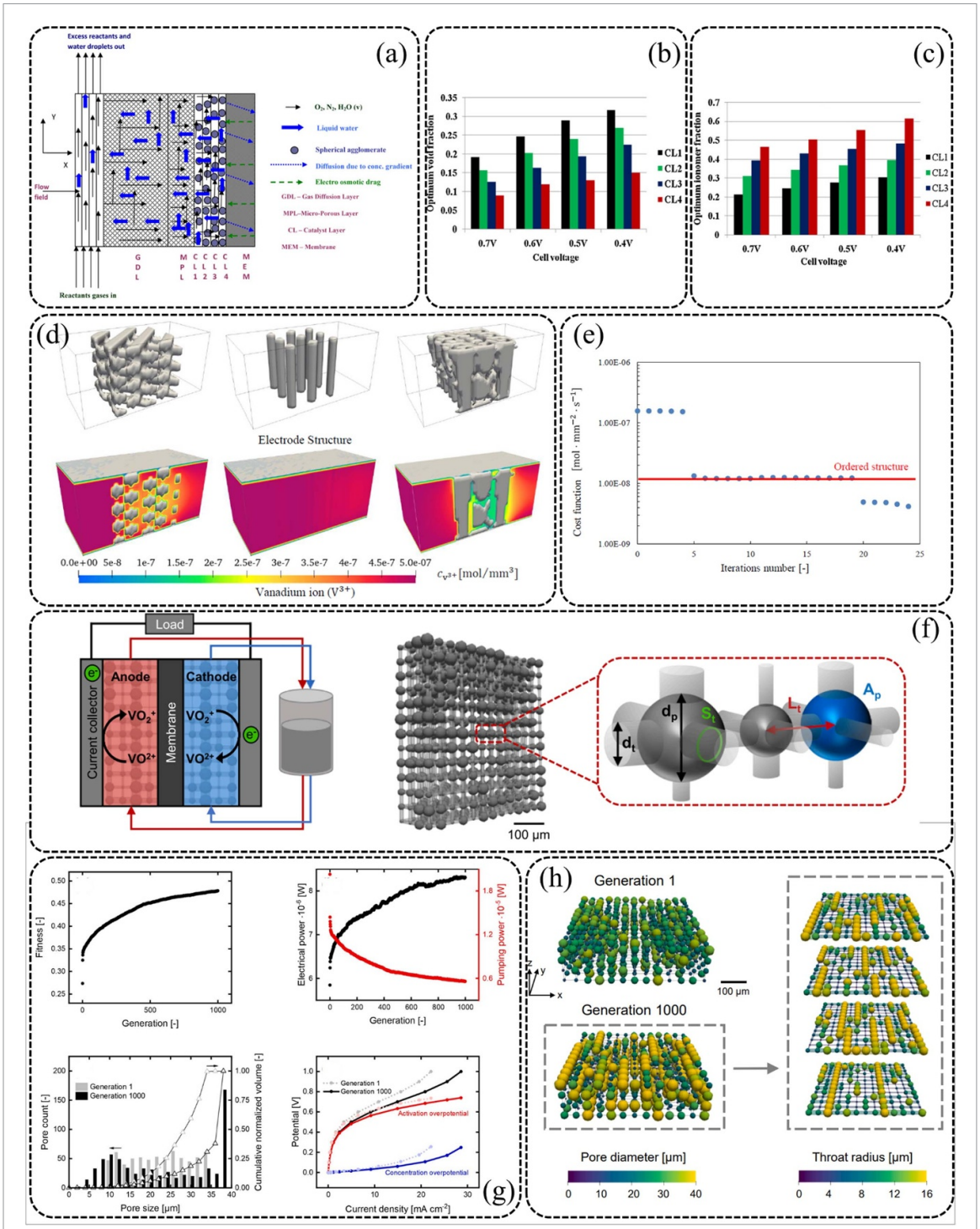

Figure 6. Examples of other notable efforts in electrode optimization (functionally graded design, LBM, and PNM). Schematics and images show (a) schematic of PEFC with multiple catalyst layer, (b) optimum void volume fraction (porosity), (c) optimum ionomer volume fraction in a multiple catalyst layer design under various terminal voltage conditions (Reprinted from [109], Copyright (2012), with permission from Elsevier); (d) results of fiber-scale simulations of electrode structure and vanadium ion concentration distribution under charging condition (optimized structure is on the right; initial structure is in the center; left is the ordered structure with a fiber diameter of 4 microns), (e) evolution of concentration flow rate at the outlet of the domain (objective function) over the optimization course in comparison to that of ordered structure (Reproduced with permission from [110]); (f) schematic representation of pore network modeling of RFB electrode, (g) evolution history of cost function, electrical power, and pumping power during the optimization as well as comparison of pore size distribution and polarization curves before and after optimization, and (h) comparison of pore network morphologies, including pore diameter and throat radius, before and after optimization Reproduced from [34] CC BY 4.0.

decrease in required pumping power as depicted in figure 6(g). The optimization also led to a 47% increase in surface area and a 42% improvement in electrochemical performance. Despite the initial motivation for using PNM to capture various phenomena at a pore level, their model uses a uniform value for the mass transfer coefficient without considering the impact of local fluid velocity. Additionally, keeping porosity

constant throughout the optimization iterations imposes an extra limitation on design freedom. The use of a cubic lattice also restricts pore movement within the design domain. This limitation was somewhat addressed in their subsequent study [115] by introducing a pore merging and splitting function. Their research highlights the importance of optimizing electrodes tailored to specific reactor designs and operating conditions. Results reveal that electrolyte kinetics and ionic conductivity can affect the final optimal design. Electrodes with a large number of tiny pores and a large surface area are more effective for kinetically slow electrolytes and high ionic conductivity. Conversely, low through-plane tortuosity and high hydraulic conductance are advantageous for kinetically active electrolytes with low ionic conductivity. A recent study [116] on multi-objective optimization of pore network morphologies in an advection–diffusion–reaction system addressed some limitations of earlier research [34, 115] by introducing a local velocity-dependent mass transfer coefficient and a morphable pore network that extends beyond a fixed cubic lattice. However, the model is limited to a first-order chemical reaction and does not account for electrochemistry-related phenomena, such as electric charge transport, species transport via electromigration, or electrochemical reactions.

5. Summary and future challenges on electrode optimization using topology optimization

TO holds the promise of crafting groundbreaking electrode structures capable of reducing material usage while enhancing the performance of EEDs. Nonetheless, several challenges loom on the horizon, including:

1. Given that topologically optimized electrode structures tend to be intricate, the fabrication process presents challenges, particularly due to the electrodes' thickness, often less than 1 mm (for PEFCs, it can be as thin as 10 μm). This issue caused many studies in this field to rely solely on mathematical computations to generate optimal designs but lack sufficient experimental validation to confirm these designs actually work well.
2. The primary drawback of this approach lies in its high computational expense, stemming from the iterative assessment of the objective function. This cost can become unaffordable, especially for complex systems with a realistic size and 3D model. To mitigate this challenge, previous research endeavors have frequently resorted to simplifying phenomena into more manageable problems or reducing the design domain or dimensionality to curtail computational expenses. Hence, incorporating all the relevant physics into the TO posed a significant challenge. In addition to that, on the mathematics side, researchers are constantly refining the algorithms used in TO. Their goal is to speed up the optimization process using one or a combination of techniques, including but not limited to multi-grid solvers, model reduction, and machine learning [117].
3. The topologically optimized electrode structure represents a mathematical solution that depends on several parameters, including the objective function, algorithmic approach, tuning parameters, and so on. However, to eliminate dependence on these factors, a robust design theory is necessary. This theory would provide a solid framework for optimizing structures without relying on specific parameters, thus bypassing the optimization process.
4. In optimization techniques like TO, a mathematical model of the system is initially developed, which is solved for evaluation of objective function(s). To ensure reliable design solutions that translate to real-world performance improvements, validating these continuum models against experimental data is crucial. While existing studies using TO for electrode design have validated their models under various operating conditions (e.g. temperature, flow rate, and relative humidity), the applicability of these models to different structural designs remains unclear. To address this, future TO applications should incorporate validation with experimental data encompassing a broader range of operational and structural variations. Specifically, the employed continuum models should accurately describe the correlation between local microstructure and overall performance [118].

In terms of fabricating topologically optimized electrodes, there is a push to experimentally demonstrate the potency of topologically optimized porous electrodes in practical applications. Currently, the fabrication of these optimized designs proves challenging due to their geometrical complexity. However, with the ongoing progress in additive manufacturing and 3D printing technologies, it is envisioned that these advancements could be leveraged to create such complex structures [119–126]. In one notable study [35], conducted by Beck and Worsley's research team, projection micro-stereolithography was employed to fabricate electrodes for electric double-layer capacitors, as illustrated in figures 7(a) and (b), resulting in observed enhancements in capacitance. Specifically, a 77% and 99% increase in capacitance was achieved for the optimized electrode compared to the control lattice electrode in numerical simulations and experiments,

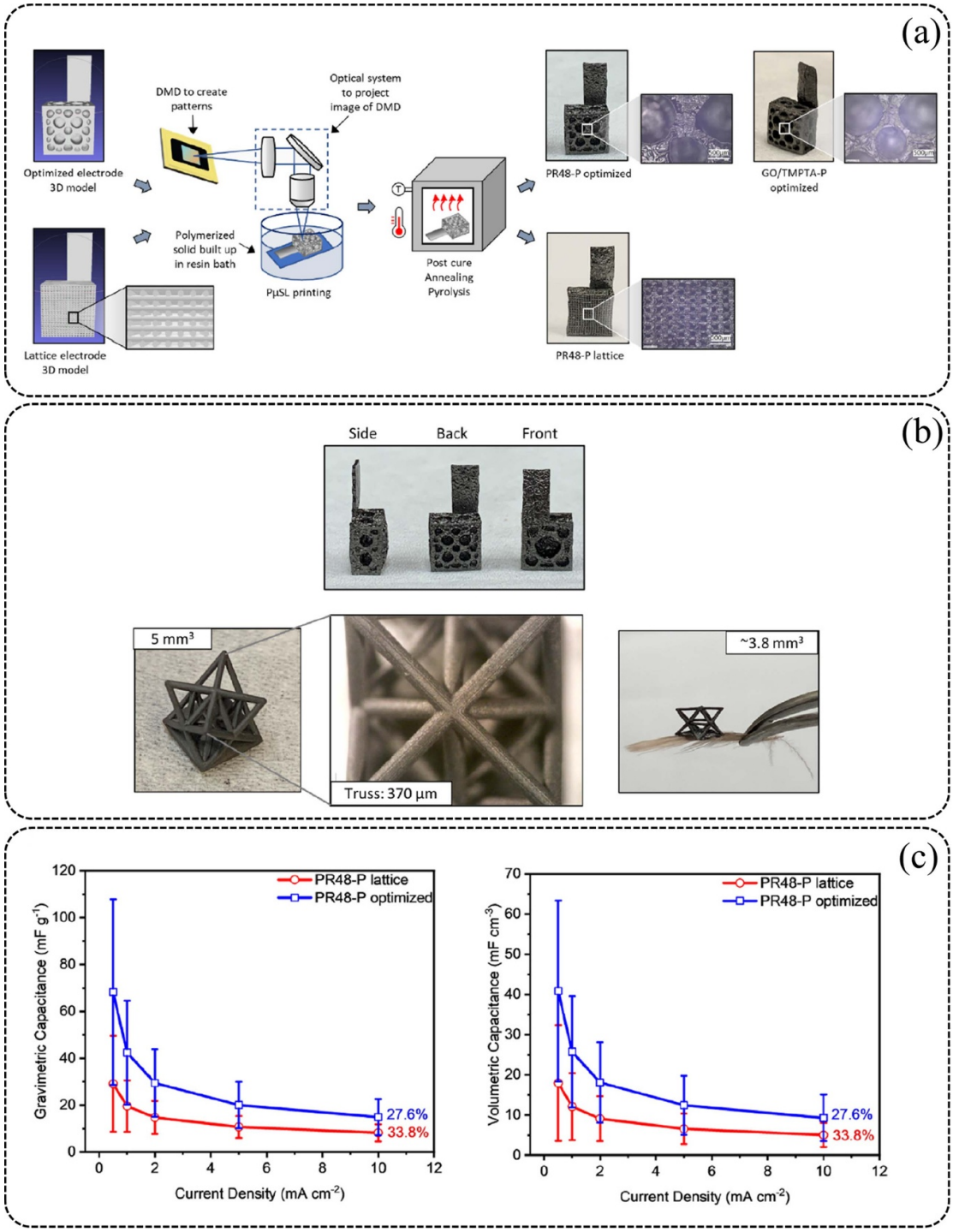

Figure 7. Examples of employing additive manufacturing to fabricate topologically optimized electrodes. Schematics and images show (a) the experimental procedure of electrode fabrication employing 3D printing technology, (b) topologically optimized and lattice porous electrodes for supercapacitor application after printing, and (c) comparison between capacitances of topologically optimized and lattice porous electrodes showing the superiority of the topologically optimized electrode over the lattice electrode (Reprinted from [35], Copyright (2023), with permission from Elsevier).

respectively (see figure 7(c)). It is essential to note that this demonstration is currently limited to applications involving electrodes with a single material. There is substantial room for researchers to delve into the fabrication of complex structures incorporating multiple materials, such as electrodes for PEFCs containing electronically conductive materials, catalysts, and ionomers. Nevertheless, this type of structure holds promise as a potential avenue for advancing the performance of electrochemical energy storage systems.

In addressing computational costs, the field has long recognized the imperative need for efficient techniques to expedite the design process. To mitigate computational expenses, effective solution schemes and innovative methodologies have been developed. In recent years, machine learning technologies,

particularly deep learning methods, have witnessed remarkable success across various applications. These methods have also been employed to alleviate the computational burden of TO by offering predictive solutions [117]. Many of these techniques are generative models, trained on optimal solutions, which can forecast solutions for similar problems under diverse conditions. The generative models depend on existing optimal designs as training data, setting them apart from conventional TO algorithms. The predictive capabilities of these models are limited by the coverage of the training datasets, necessitating the consideration of new datasets and networks to accommodate diverse domain geometries or constraints. Motivated by these limitations, Deng *et al* [127] recently proposed the Self-directed Online Learning Optimization (SOLO) algorithm to significantly expedite the TO process. This algorithm approximates the original costly-to-calculate function by replacing it with a deep neural network (DNN), which learns and maps designs to objectives as a surrogate model. Based on the DNN's optimal predictions, a small amount of training data is dynamically generated. As the algorithm converges, the DNN adapts to the new training data, providing improved predictions in the area of interest. SOLO was tested on four types of simple problems—truss optimization, heat transfer enhancement, fluid-structure optimization, and compliance minimization—and outperformed state-of-the-art algorithms. It substantially reduced computational time by 2–5 orders of magnitude compared to directly applying gradient-free heuristic optimization. In the context of utilizing TO to discover groundbreaking electrode structures in EEDs, the integration of machine learning-assisted algorithms is particularly vital. Electrodes pose a complex 3D problem involving multiple non-linear coupled partial differential equations (mass, electronic charge, and ionic charge transports as well as electrochemical rate process). Therefore, employing machine learning techniques can significantly enhance the efficiency and effectiveness of TO processes in this domain.

Various objective functions, algorithmic approaches, and tuning parameters yield diverse optimal solutions in TO. To ascertain whether a solution obtained is globally optimal, robust theoretical frameworks are essential. Recently, efforts have been made to link entropy generation to topologically optimized structures, proposing that the optimized structure should minimize entropy generation. Recent findings have indeed demonstrated a connection between entropy generation and optimized structures [33, 97–99]. However, while entropy generation analysis offers valuable insights into electrode design, it falls short of becoming a comprehensive design theory capable of guiding the creation of optimal electrode structures. Research in establishing a theory to design electrodes for high-performance EEDs is pivotal for advancing energy storage and conversion technologies. While the ultimate goal is to develop a comprehensive design theory that obviates the need for TO, current TO outcomes can serve as valuable benchmarks and represent the best available designs for specific applications. The key challenge now is to establish a robust theoretical framework capable of generating these optimal designs without relying on numerical techniques like TO. Such a theory would provide researchers with a systematic approach to electrode design, enhancing the efficiency and effectiveness of EEDs.

In summary, TO for the design of porous electrodes in electrochemical energy storage and conversion systems represents a new emerging research direction. Topologically optimized electrodes possess the potential to overcome longstanding barriers in efficiency, cost, and performance. By reviewing studies published in recent years, we highlight the main challenges spanning from computer-aided design through TO to the practical realization of these innovative designs. Furthermore, by exploring the intersection of natural design principles and engineering innovation, this paper inspires future research directions that not only enhance technological capabilities but also deepen our understanding of complex systems in nature. The synthesis of cutting-edge research presented in this review is expected to stimulate fruitful discussions and inspire new avenues of inquiry, thereby contributing to the advancement of the global transition towards a sustainable energy future.

Data availability statement

The data cannot be made publicly available upon publication because they are not available in a format that is sufficiently accessible or reusable by other researchers. The data that support the findings of this study are available upon reasonable request from the authors.

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ORCID iDs

Mehrzad Alizadeh https://orcid.org/0000-0001-5086-1683
Patcharawat Charoen-Amornkitt https://orcid.org/0000-0001-6382-3715
Shohji Tsushima https://orcid.org/0000-0001-8935-0135

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