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Review

Computational design and optimization of fuel cells and fuel cell systems: A review

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ABSTRACT

The design of fuel cells is a challenging endeavour due to the multitude of physical phenomena that need to be simultaneously optimized in order to achieve proper fuel cell operation. Fuel cell design is a multi-objective, multi-variable problem. In order to design fuel cells by computational design, a mathematical formulation of the design problem needs to be developed. The problem can then be solved using numerical optimization algorithms and a computational fuel cell model. In the past decade, the fuel cell community has gained momentum in the area of numerical design. In this article, research aimed at using numerical optimization to design fuel cells and fuel cell systems is reviewed. The review discusses the strengths, limitations, advantages, and disadvantages of optimization formulations and numerical optimization algorithms, and insight obtained from previous studies.

Contents

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Nomenclature

$\dot{V}$	volumetric flow rate [$m^3\,s^{-1}$]
i	current density [$A\,cm^{-2}$]
m_{Pt}	platinum loading in the catalyst layer [$mg\,cm^{-2}$]
P	power density [$W\,cm^{-2}$]
p	pressure [Pa]
$Pt\|C$	platinum to carbon weight ratio of the catalyst support
RH	relative humidity [%]
T	temperature [K]
t_i	thickness of layer i [cm]
V_{cell}	cell voltage [V]
w_{cc}	width of the current collector [cm]
w_{ch}	width of the gas inlet channel [cm]

Greek letters

ϵ_N^{cl}	electrolyte phase volume fraction in the CL
ϵ_S^{cl}	solid phase volume fraction in the CL
ϵ_V^{cl}	porosity or void volume fraction in the CL
ϵ_V^{gdl}	Porosity or void volume fraction in the GDL
ϵ_{agg}	electrolyte volume fraction in the agglomerate
η	efficiency [%]
λ_a	gas stoichiometry at the anode
λ_c	gas stoichiometry at the cathode
λ_N	membrane hydration
ϕ_N	electric potential in the electrolyte phase [V]
ϕ_S	electric potential in the solid phase [V]

1. Introduction

Design of fuel cells is a challenging endeavor due to: (a) the multitude of physical phenomena that need to be simultaneously optimized to achieve proper fuel cell operation, (b) the multitude of design objectives that need to be optimized, (c) a lack of accurate fuel cell models, and, (d) the amount of computational resources necessary to solve the governing equations of a fuel cell. For example, in order to optimize a membrane-electrode-assembly (MEA), the following objectives need to be achieved: (a) minimize cost, i.e. by reducing platinum loading in the electrodes, (b) maximize the performance of the MEA (in terms of either power density or current density for a given voltage range), (c) minimize the effects of contaminants in the anode and cathode, and (d) minimize electrode and membrane thermal, chemical and mechanical degradation. In order to achieve these goals, several parameters need to be optimized, such as: (a) the geometry of the gas diffusion layer (GDL), micro-porous layer (MPL), catalyst layer (CL) and proton exchange membrane (PEM), and (b) the composition and microstructure of GDL, MPL, CL and PEM, e.g., porosity, platinum loading, and ionomer loading. There is a trade-off between objectives; therefore, all objectives need to be optimized simultaneously. For example, minimizing platinum loading will reduce cost but it will most likely also reduce fuel cell performance. Similarly, there is a trade-off between the design variables, e.g., increasing CL porosity will lead to improved mass transport but it will also result in a decrease in charge transport due to a reduction in either ion or electron conductive materials. As a result of these trade-offs, all design parameters and objectives need to be optimized simultaneously.

Due to the large number of design objectives and design variables, trial-and-error approaches to fuel cell design are time-consuming and, in most cases, lead to a sub-optimal design. The main reason for obtaining sub-optimal designs is that the number of possible designs that need to be evaluated in order to obtain the optimal solution increases extremely quickly with the number of design variables. As an example, for a design with ten design variables and six possibilities per design variable, the number of possible designs is 10^6. It is obvious that it is impossible to manually evaluate all designs. Therefore, design studies based on trial-and-error approaches are usually limited to one objective and a handful of design variables. For example, experimental studies in electrode optimization and flow channel design have been mainly concerned with optimizing performance (i.e. current density), while ignoring any of the other design objectives such as cost, pressure drop in the channel, and robustness to contaminants and degradation. Experimental studies aimed at obtaining the electrode structure that achieves maximum performance have only studied the effect of either one design parameter (e.g., Nafion loading [1–4] or platinum loading [5]), or at most two design parameters (e.g., platinum and Nafion loading [6]). The result is that the coupling between all design parameters is not properly captured and a sub-optimal design is obtained.

With the proliferation of fuel cell analysis computer models, it is now possible to use computer models to design fuel cells. The next logical step is to automate the design process. Design problems with a large number of design objectives and variables are common in structural, aerospace, and automotive engineering. In these industries, numerical optimization is commonly used to solve these problems [7]. Numerical optimization has been a very active area of research since the 1960s, and many optimization formulations [8] and optimization algorithms have appeared in the literature [7,9,10]. Computational optimization has been used in many applications, for example in obtaining the optimal shape of airfoils [11], in simultaneously optimizing the structure and shape of aircraft wings [12], and in finding the optimal topology of automotive parts and micro-electro-mechanical systems (MEMS) [13].

Computational optimization is based on coupling a mathematical algorithm with a computational analysis tool in order to efficiently search for an optimal design. Using the optimization algorithm, only a few designs need to be evaluated, thereby substantially reducing the necessary computational time to achieve an optimal design. Further, in many cases, the optimization process can guarantee that the design is an optimal one. Using computational optimization liberates engineers from the tedious task of performing parameteric studies in order to improve a new design, thereby enabling them to concentrate on developing innovative designs instead of fine-tuning existing ones.

In the area of fuel cell design, the application of numerical optimization is scarce. A total of approximately 49 articles have appeared in the literature in the areas of: (a) flow field design, (b) electrode design, (c) operating conditions optimization and, (d) fuel cell assembly optimization. All these publications have appeared within the past decade, with most in the past 5 years, indicating the growing interest in this field of research.

This article reviews only previous work on fuel cell and fuel cell system design using numerical optimization. Only stand-alone fuel cell system optimization will be reviewed; hybrid power systems where the focus is not in fuel cell design are not included. Studies in the area of computational fuel cell design that are based on parameter estimation, such as references [14–17], are also not included. The article is structured as follows. First, a review of optimization algorithms is presented in Section 2. The review is focused on algorithms to solve single-objective and multi-objective constrained optimization problems because these are the problems most commonly encountered in fuel cell design. Section 3 presents the different optimization approaches that have been used in the literature to optimize channel geometry, electrode geometry and composition, operating conditions of single cells and the design of fuel cell stacks. Section 4 describes research on design and opti-

mization of fuel cell system. Section 5 provides conclusions and recommendations based on previous work.

2. Numerical optimization

2.1. Problem formulation

Fuel cell design and optimization usually involves solving a multi-objective, nonlinear constrained optimization problem of the form

$$\begin{array}{lllll} \text{minimize} & f(\mathbf{x}) & & & \text{(a)} \\ \text{w.r.t.} & x_k & \text{for} & k = 1, 2, \ldots, n & \text{(b)} \\ \text{subject to :} & h_i(\mathbf{x}) = 0 & \text{for} & i = 1, 2, \ldots, p & \text{(c)} \\ & g_j(\mathbf{x}) \leq 0 & \text{for} & j = 1, 2, \ldots, q & \text{(d)} \\ & \mathbf{x}_L \leq \mathbf{x} \leq \mathbf{x}_U & & & \text{(e)} \end{array} \quad (1)$$

where the function to be minimized, $f(\mathbf{x})$, is known as the objective function. In fuel cell design, there are several objectives that should be optimized, such as cost, performance at several operating points, and durability. The objective function is one or a combination of these objectives that depends on a set of variables, $\mathbf{x}$, which can take arbitrary values. These variables are known as the design variables. Design variables in fuel cell design are parameters such as platinum loading, amount of ionomer in the catalyst layer and the height and width of a gas channel. The design variables are usually bounded. In Eq. (1), $\mathbf{x}_L$ and $\mathbf{x}_U$ represent the vector of lower and upper bounds for the design variables. The aim of the optimization algorithm is to obtain the value of the design parameters, $\mathbf{x}$, that makes the objective function minimal. This point is known as the solution of the optimization problem and is represented by $\mathbf{x}^*$. It is important to notice that minimizing a function, $f(\mathbf{x})$, is equivalent to maximizing the function $m(\mathbf{x}) = -f(\mathbf{x})$. Most engineering designs also need to satisfy several physical and manufacturing constraints, therefore design constraints are also included in the design problem such as a maximum pressure drop in the channel and a minimum porosity. In Eq. (1), $h_i(\mathbf{x}) = 0 \text{for} i = 1, 2, \ldots, p$ are the equality constraints and $g_j(\mathbf{x}) \leq 0 \text{for} j = 1, 2, \ldots, q$ are the inequality constraints. It is assumed that functions $f(\mathbf{x})$, $\mathbf{h}(\mathbf{x})$, and $\mathbf{g}(\mathbf{x})$ are nonlinear, continuous and have continuous first and second order derivatives.

2.2. Nonlinear constraint optimization algorithms

Nonlinear constrained optimization problems involve the search for a minimum of a nonlinear objective function subject to a set of nonlinear constraints. It is common for a nonlinear optimization problem to have multiple extrema. Due to this difficulty, two different approaches have emerged in the area of nonlinear constraint optimization: local methods and global methods. Local methods aim to obtain a local minimum, and they cannot guarantee that the minimum obtained is the absolute minimum. These methods are usually first-order methods, i.e. they require information about the gradient of the objective function and the constraints. On the other hand, global methods aim to obtain the absolute minimum of the function. They do not need any information about the gradient, and they are mostly based on stochastic procedures.

Local constrained methods can be classified into sequential methods and transformation-based methods. Sequential methods aim to solve the nonlinear constrained problem by iteratively solving a simpler constrained optimization problem. The most commonly used local sequential methods are: the method of feasible directions (MFD) and modified method of feasible directions (MMFD) [7,10,18], sequential linear programming (SLP) [7,10,19,20], sequential quadratic programming (SQP) [21,22], and surrogate-based optimization methods [23–27].

Local transformation-based methods transform the original nonlinear optimization problem into an unconstrained optimization problem by adding a penalty function to the objective function. Once the constrained problem has been transformed into an unconstrained problem, any unconstrained optimization algorithm can be used to solve the transformed problem. The most commonly used local transformation-based methods are: penalty methods [7,10] and augmented Lagrangian methods [7,10,18]. All penalty methods have a main drawback; due to the introduced penalty, the objective function becomes highly nonlinear and this makes it difficult for the unconstrained methods to obtain the minimum.

It is important to note that, although local methods do not aim for the global optima, they can be used to obtain said global optima. Several methods can be used to continue searching once a local minimum has been obtained, thereby enabling the identification of all local minima. Some of these methods, based on a stochastic approach, are: random multi-start methods [28,29] and ant colony searches [30]. Some methods use a deterministic approach to find the global optimum. For example, the local-minimum penalty method [31] includes a penalty to the objective function if the algorithm tends to go to an already known local minima.

Global methods can be classified into two groups: direct or transformation-based methods. Direct methods include, for example, covering methods [32] and pure random searches. The main drawback of direct methods is that they require a large number of function evaluations and are therefore computationally expensive.

Global transformation-based methods first transform the original problem into an unconstrained problem. Then, global unconstrained techniques are used to obtain the global minima. Commonly used unconstrained global methods are: genetic algorithms (GA) [33], evolutionary algorithms [34] and simulated annealing [35]. These methods have the same drawback as the global direct methods; they require a large number of objective function evaluations. As a result, the computational requirements for these methods become prohibitive when the evaluation of the objective function and constraints is time consuming.

2.3. Multi-objective optimization algorithms

In fuel cell design, the designer would like to optimize several coupled design objectives such as the minimization of cost, maximization of performance and the maximization of durability. In order to optimize several objectives simultaneously, a multi-objective optimization problem needs to be formulated. A mathematical formulation of such a problem is given by

$$\begin{array}{lll} \text{minimize} & \mathbf{J}(\mathbf{x}) = & [J_1, J_2, \ldots, J_n]^T \\ \text{w.r.t.} & x_k & \text{for} \quad k = 1, 2, \ldots, n \\ \text{subject to :} & h_i(\mathbf{x}) = 0 & \text{for} \quad i = 1, 2, \ldots, p \\ & g_j(\mathbf{x}) \leq 0 & \text{for} \quad j = 1, 2, \ldots, q \\ & \mathbf{x}_L \leq \mathbf{x} \leq \mathbf{x}_U & \end{array} \quad (2)$$

where J_i is one of the objectives to be minimized, $\mathbf{J}(\mathbf{x})$ is the vector of objectives and the other constraints have the same meaning as in Eq. (1).

The scalar concept of optimality does not apply directly to the multi-objective problem because there are more than one optimal solutions depending on the importance of each objective. A useful notion is that of the *Pareto optimality*. A design, $\mathbf{x}$, is a *Pareto optimal solution* for the problem (2), if all other feasible designs have a higher value for at least one of the objective functions J_i, or else have the same value for all objectives [36–38]. Using the definition of Pareto optimality, there are many Pareto optimal solutions, $\mathbf{x}^*$. Since all Pareto optimal solutions are good solutions, the most appropriate solution will depend only upon the trade-offs between objectives; therefore, it is the responsibility of the designer to choose the most appropriate solution. The *Pareto front* is a set that contains the objectives of all optimal solutions. It is

sometimes desirable to obtain the complete set of Pareto optimal solutions, from which the designer may then choose the most appropriate design.

There are a variety of methods for solving multi-objective problems and for obtaining the set of Pareto optimal solutions [36,38–42]. One of the most widely used methods for multi-objective optimization is the weighted sum method [40]. In this method, the multiple objectives are transformed into a single objective function by multiplying each objective by a weighting factor and summing up all contributions. Each single set of weights determines one particular Pareto optimal solution. The weighted sum method is easy to implement and it can easily be understood; however, it has two drawbacks: (1) a uniform spread of weight parameters rarely produces a uniform spread of points on the Pareto set; (2) non-convex parts of the Pareto set cannot be obtained [43].

In the literature, only a handful of articles have addressed the multi-objective nature of fuel cell design. As an example, Secanell et al. [44] optimized both performance and platinum loading. To obtain the Pareto set they used a weighted sum method. The method was shown to provide good results. Chen et al. [45] optimizing fuel cell operating conditions in order to minimize the capital and operating costs of the fuel cell. The multi-objective function included three objectives: (a) minimize the annualized cost of the cell, (b) minimize the fuel costs, and (c) maximize the credits for the exhaust hydrogen.

2.4. Sensitivity analysis

Sensitivity analysis is concerned with obtaining the change of a certain output variable with respect to an input variable, i.e. gradients or sensitivities. In the case of optimization, sensitivity analysis is used to obtain the derivatives of the objective function and constraints with respect to the design variables. These values are needed by the gradient based optimization algorithms discussed in Section 2.2. In some cases, gradient based methods might also need the second derivatives, i.e. the Hessians. These values can be computed analytically or they can be approximated. Quasi-Newton methods, such as the Broyden–Fletcher–Goldfarb–Shanno (BFGS) method, use an approximation of the Hessians to solve the optimization problem.

In the literature, several methods have been suggested to compute the gradients of physical properties with respect to the design variables. This include: (a) finite difference differenciation, (b) complex-step differentiation, (c) automatic differentiation, and (d) analytical differentiation. Finite difference differentiation uses a Taylor series expansion of a function around a point, $\mathbf{x}_0$, to obtain an approximation of the gradient. First-order finite difference differentiation needs $n+1$ function evaluations to compute the gradient of a function, with n being the number of independent variables. First-order forward difference is easy to implement and is computationally more efficient than complex-step differentiation and automatic differentiation methods [46,47]. However, forward-difference is also the most inaccurate of all the methods described above and it is subject to the step-size dilemma, i.e. the error is proportional to the step size but it is very difficult to find the appropriate step size due to: (a) the first order approximation, and (b) numerical errors for small step sizes [46,47].

Complex-differentiation solves the step-size dilemma encountered in the finite-difference method by using a complex step to compute the gradients [46,48,49]. The approximation is second order. The number of function evaluations necessary to obtain the gradient is still $n+1$ where n is the number of independent variables of the function. In order to obtain the gradients using complex-step differentiation, the source code of the analysis program has to be changed so that all the real variables become complex variables.

Automatic differentiation (AD) is based on successive application of the chain rule to each operation performed in the analysis computer code [50,51]. Since the structure of a computer code is composed of a successive set of arithmetic operations used to compute the value of a function, successive application of the chain rule to each one of the operations in the code will result in the exact (to machine precision) desired derivatives. In order to transform a code into a forward or reverse automatic differentiation code, there are several programs that precompile the original code and transform it into an AD code. Some of the codes that can be used to transform either FORTRAN or C++ source codes to AD codes are: ADIFOR, IMAS, Tapenade, OPTIMA90 and the Trilinos/Sacado AD library.

Finally, analytical differentiation consists of deriving the analytical expressions for the sensitivities and introducing them to the original analysis code. These methods are the most efficient and accurate; however, they are also the most difficult and time consuming to implement. There are two methods used to compute the sensitivities analytically: direct methods and adjoint methods. Using the direct method, the computations necessary to obtain the gradients of M functions with respect to N design variables is the solution of N linear systems of equations of similar size as the original problem. The adjoint method eliminates the dependence of the gradient computations on the number of design variables [52]. Therefore, the computations necessary to obtain the gradient of M functions with respect to N design variables is the solution of M linear systems of equations of similar size as the original problem. For fuel cell design problems involving a large number of design variables and a small number of objectives and constraints, such as for flow channel shape optimization, the adjoint method is the most effective method for performing sensitivity analysis.

Developments in sensitivity analysis for fuel cell applications are scarce. Sensitivity analysis to perform optimization using gradient-based methods has mainly been based on finite difference differentiation [53–64]. However, forward-difference differentiation has the two major drawbacks described previously. In the fuel cell literature, only three groups have developed an alternative to the numerical sensitivities. Kapadia et al. [65,66] developed a three-dimensional numerical model of a solid oxide fuel cell capable of obtaining the sensitivities of several objectives with respect to the material properties of the solid oxide fuel cell. To compute the sensitivities, both an adjoint and direct method are implement. Carnes and Djilali [67] developed a one-dimensional fuel cell model capable of obtaining the analytical sensitivity of the current density with respect to several physical parameters using the direct method. Secanell et al. [68–70,44] developed a two-dimensional numerical model of a polymer electrolyte fuel cell that is capable of obtaining the analytical sensitivities of the current density using the direct method.

3. Fuel cell design and optimization

The following section will review the published articles in single cell and stack design for fuel cells. Much of the modeling and optimization efforts in this area are concentrated on low-temperature fuel cells. Section 3.1 includes studies using polymer electrolyte (or proton-exchange) membrane fuel cells (PEFCs), both hydrogen or direct methanol fuel cells (DMFC). The limited number of optimization studies using high-temperature fuel cells, namely solid oxide fuel cells (SOFC), will be discussed in Section 3.2

3.1. Low-temperature fuel cells

Efforts on computational design and optimization of PEFCs have been focused on two areas: (a) design of flow fields, and (b) design of electrodes. Usually, computational design studies that have focused

on the flow field have used a very simple model for the electrode in order to reduce the computational expense of the analysis program. Similarly, studies that have focused on optimizing the electrode structure and composition have usually neglected the convective transport in the channel and porous media.

3.1.1. Flow field optimization

The first research efforts in the area of fuel cell flow field design and optimization were published in 2004. Grujicic et al. presented several articles [54–56] concerning the optimization of the geometric parameters in the cathode of an interdigitated fuel cell. In Ref. [54], the authors optimized the current density of an interdigitated fuel cell at a cell voltage of 0.7 V. The design variables were: (a) the inlet cathode oxygen pressure (p_c^{in}); (b) the cathode GDL thickness ($t_{GDL,c}$); (c) the width of the gas channel plus current collector ($w_c = w_{cc,c} + w_{ch,c}$), and (d) the fraction of the current collector ($\Lambda_c = w_{cc,c}/(w_{cc,c} + w_{ch,c})$). The model used to perform the optimization was a two-dimensional model of an MEA (including GDL, PEM and a zero-thickness CL model). The model used Fick's law and Darcy's law to account for transport of fuel and reactants and Ohm's law for the transport of electrons through the GDL. The PEM model only accounted for the transport of charge and used a constant membrane conductivity. The governing equations were implemented and solved using COMSOL Multiphysics. The optimization problem of maximizing the current density subject to bounds for all design variables was solved using *fmincon*, the sequential quadratic programming algorithm in MATLAB. All design variables reach the bounds in the optimal design. Inlet pressure and current collector width reach their respective upper limits while the cathode thickness and fraction of the current collector reach the lower limits. Therefore, no trade-off is observed in this optimization problem. In addition to obtaining the optimal design, Grujicic et al. also performed a detailed design robustness analysis with respect to: (a) reference oxygen concentration, (b) cathode/membrane equilibrium potential, (c) active layer thickness, (d) gas diffusive coefficient inside the agglomerate, and (e) cathodic exchange current density. It was noted that the optimal design is unaffected by a ±10% variation in these parameters.

In Ref. [55], Grujicic et al. again aim at optimizing the geometry of the gas distributors in order to achieve a maximum current density. In this case, a three-dimensional model of the cathode of an interdigitated fuel cell is used in order to obtain the optimal value for the following design variables: (a) the GDL thickness ($t_{GDL,c}$); (b) the height of the interdigitated air distributor channel ($h_{ch,c}$); and, (c) the width of the air distributor channel ($w_{ch,c}$). The three-dimensional isothermal, single-phase model used for optimization includes the cathode gas distribution channels and the GDL. At the GDL–CL interface, the reactions are accounted for via a flux boundary condition. The governing equations for the channels and cathode electrode are a mass continuity equation, the Navier–Stokes equation and the Maxwell–Stefan equation for oxygen and water. No justification is given for using the Navier–Stokes equation without any correction such as the Darcy damping force in the cathode electrode, a porous media. The optimization results show that the channel height and width reach their upper bounds. The optimal cathode GDL thickness does not reach the bounds and the optimal value is 37 μm. Comparing this result to their previous publication [54], the thickness does not reach the bounds because the bounds have been reduced substantially from the previous study.

In Ref. [56], the current density of an interdigitated flow field fuel cell is again optimized with respect to the same design variables as in Grujicic et al.'s previous work [54] with the exception of the total pressure which has been removed. The fuel cell model used in this case is similar to the model used to simulate the cathode in reference [54]; however, the cathode model is extended to include a mass balance equation for water vapour and an equation for saturation. The optimal solution has a GDL thickness of 25 μm and the width of the gas channel and the fraction of the current collector both reach its lower bound. The contrasting results between Refs. [54] and [56], where the current collector width reaches its upper and lower bound, respectively, are not discussed but are likely due to the effect of saturation.

More recently, Lin et al. [57] also analyzed the optimal channel width ratio using numerical optimization. They optimized the power density of a PEMFC cathode electrode at an overpotential of 0.25 V (cell voltage is not reported) using an in-house PEMFC electrode model and a gradient-based optimization algorithm (simplified conjugate gradient method; SCGM). The optimization design variables are: (a) the CL porosity (ϵ_V^{CL}); (b) GDL porosity (ϵ_V^{GDL}), and (c) the gas channel width ratio ($\lambda = w_{ch}/w_{cc}$ where w_{ch} is the width of the channel and w_{cc} is the width of the current collector). The fuel cell electrode model solved the transport of oxygen through the CL and GDL and the transport of electrons through the CL, GDL and bipolar plate. The electrolyte potential was not considered and instead a fixed overpotential was used to compute the reaction rate of the electrochemical reaction using a Tafel equation. No constraints were used in the optimization process. The results showed an optimal channel width ratio of 0.54 and GDL and CL porosites of 0.6 and 0.3 respectively.

Cheng et al. [58] integrated a commercial CFD package (CFD-ACE+) with a gradient-based optimization algorithm (simplified conjugate gradient method; SCGM). The sensitivity analysis was performed numerically, most likely using forward differences. The objective is to maximize power density at a cell voltage of 0.7 V. Since voltage is fixed, this is equivalent to optimizing the cell current density at 0.7 V. The design variables are: (a) the gas channel width fraction (λ); (b) the gas channel height (h_{ch}); and, (c) GDL thickness (t_{GDL}). The solution of the optimization problem took 30 iterations and 60 h of CPU time in a personal computer. The objective function used is the inverse of the power density, so a minimum can be calculated by the optimization algorithm. It is unclear why this approach is taken, as minimizing the negative current has the same effect without introducing further nonlinearity to the design space. It may be possible to reduce the solution time by simplifying the optimization formulation and gradient calculations.

In 2009, Huang et al. [59,60] and Wang et al. [61] presented the first studies aimed at changing the geometry of the channel in the direction of the flow. These articles provide the first real attempt at flow field shape design optimization. Huang and Lin [59] solved an inverse problem in order to redesign the channel height of the outlet section of a triple serpentine PEMFC in three-dimensions. The standard PEMFC module provided by CFD-ACE+ is used to model the transport phenomena inside the fuel cell. The channel section is parameterized using a second order B-spline with three control points, which are used as design parameters. The objective of the study is to redesign the channel height in order to increase the current density under the redesigned section by 15% with respect of the original design at a cell voltage of 0.3V. The optimization problem can be stated as

$$\begin{aligned} &\text{Minimize} \quad J(B_j) = \sum_{i=1}^{M} (I_{c,i}(B_j) - I_{d,i})^2 \\ &\text{w.r.t.} \qquad B_j \end{aligned} \tag{3}$$

where $i = 1, \ldots, M$ is the number of points at which the current density is evaluated, $j = 1, \ldots, 3$ is the number of control points, $I_{c,i}(B_j)$ is the current density of the redesigned cell at point i and $I_{d,i}$ is the desired current density at point i. Two cases are studied: (a) redesign of the last 20 mm of the channel outlet, and (b) redesign of the last 40 mm of the channel outlet. The increase in current density results in an increase in the cell current density of 1.4% and 3.3% for cases (a) and (b), respectively. The optimization problem is

solved using the Levenberg–Marquardt method, a gradient-based minimization method. The sensitivities of the objective function are computed as follows:

$$\frac{dJ}{dB} = \sum_{i=1}^{M}\sum_{j=1}^{P} 2(I_{c,i}(B_j) - I_{d,i})\frac{dI_{c,i}}{dB_j} \quad (4)$$

where $dI_{c,i}/dB_j$ is computed by numerical differentiation. To compute $dI_{c,i}/dB_j$, each design variable (control point) is perturbed and a new CFD analysis is performed. Therefore, the computation of the derivatives requires one additional CFD computation per design variable. The CPU time on an Intel Dual-Core 1.8 GHz processor is about 109 h and 65 h for cases (a) and (b), respectively, and is obtained after only 5 and 3 iterations. The large amount of computational time illustrates the need for an efficient method to compute the sensitivities of the objective function, e.g. analytical sensitivities.

The computational optimization results in Ref. [59] were validated experimentally. The results show that performance is increased as predicted by the new design. At 0.3 V, the total density is increased from 55.00 A to 55.75 A and 56.76 A for cases (a) and (b), respectively. This improvement illustrates the advantage of using numerical optimization. It is important to notice that further improvements are possible. In this study, the target was to improve the local current density by 15% in the redesigned sections. The problem could be reformulated to maximize the current density in the redesigned section leading to the maximum possible performance for a fuel cell with a redesigned fuel cell outlet.

In Ref. [60], Huang et al. applied the inverse design technique described above to redesign a section of a single straight channel. Three cases are studied in which the channel length (as measured from the outlet) are redesigned: (a) 20 mm, (b) 15 mm, and (c) 10 mm. The objective was to increase the current density at the redesigned section of the channel by either 20% or 30% at a cell voltage of either 0.4 V or 0.7 V. Only for a cell voltage of 0.4 V could a 30% improvement be achieved. The design variables are four control points of a B-spline that controls the height of the channel. The PEMFC model used is the same as that used in [59], i.e. the standard PEMFC module provided by CFD-ACE+. The optimization results are obtained after 3–4 days of CPU time. The optimal geometry of the redesigned section is similar to that presented for the triple-serpentine channel in [59]. The redesigned section of the channel shows a sudden drop in height and then a near-constant height is maintained. The sudden reduction in height helps to redirect some of the flow in the channel towards the GDL, removing excess water and increasing the oxygen concentration. Unfortunately, the new design presents a sharp increase in pressure drop. Even though this is not discussed in the article, the increased presure drop could lead to an increase in the sizing of the air compressor, thereby reducing system efficiency and increasing the cost of the system.

Wang et al. [61] maximized the power density, at a voltage of 0.4 V, of a PEMFC with a single serpentine flow field with four bends by changing the height of the bends and width of each section of the serpentine channel. The geometry of the anode and cathode flow fields is considered to be the same. Eight design variables were used, five design variables describing the height of the serpentine bends and three describing the width of the serpentine channel sections. The width of the serpentine channel ribs was maintained constant. The optimization problem was solved using the simplified conjugate gradient method (SCGM). After approximately 35 iterations, the optimal solution was obtained. CPU time to obtain the solution was not reported. The optimal design achieves an increase in power density of 22.5% with respect to the base design (a serpentine channel with uniform cross section channels of 1 mm × 1 mm channels). The optimal flow field has a converging inlet section (H_{in} = 1 mm and H_{out} = 0.25 mm) and a diverging outlet section in the serpentine channel (H_{in} = 0.1 mm and H_{out} = 0.69 mm). The width of the outlet section is also reduced with respect to the base design (0.44 mm vs. 1 mm), thereby reducing the cross-sectional area and increasing the speed of the fluid in this zone. The sections of the serpentine channel in between the inlet and the outlet sections present a reduced height with respect to the base case (0.35–0.1 mm vs. 1 mm) and a variable width towards the outlet (1.19, 1.52, and 0.85 mm). The optimal design increases performance at the expense of a large increase in pressure drop in the channel. The trade-offs between increased performance and increased pressure drop are not discussed. An increase in pressure drop will result in an increase of the power demands to the auxiliary systems and might result in a reduction of the power density of the complete fuel cell system.

Optimization of the pressure drop in gas channels was the motivation of the recent study by Zhang et al [71]. Serpentine channels provide a more uniform distribution of reactants than parallel channels, but they usually result in larger pressure drops. Zhang et al. [71] used numerical optimization in order to design an 11-channel Z-type bipolar plate that has a uniform flow distribution. In order to predict flow distribution, i.e. the velocity in the manifold channel and individual channels, the research group developed an analytical model to calculate the flow distribution in parallel channels. The model is based on using an analogy between fluid flow in the channels and current in electrical circuits. The analytical model was validated by comparison of the analytical results with predictions from CFD-ACE+ and good agreement was observed. The design problem was stated as obtaining the width of the channels and land area for each one of the channels such that the following objective is minimized

$$f(\mathbf{x}) = \frac{1}{N\bar{v}}\sqrt{\sum_{i=1}^{N}(v_i - \bar{v})^2} \quad (5)$$

where v_i is the mean velocity of the reactants in channel i, $\bar{v}$ is the mean velocity in the plate and N is the number of channels (11 in the article). The objective is the weighted, least-square difference between the average bipolar plate velocity and the individual channels such that if the value is zero, all channels would have the same velocity. The design variables are constrained to practical manufacturing sizes (i.e., the channels and ribs should be larger than 0.8 mm and smaller than 1.5 mm) and symmetry is maintained in the bipolar plates by equality constraints for what Zhang et al. [71] cite as "engineering purposes." In the opinion of the authors, equality constraints lead to optimization problems that are difficult to solve; therefore, they should be eliminated when possible. An alternative approach would have been to impose the constraints in the analysis part of the problem instead of in the optimization formulation. The obtained design contains channels with variable width with the smaller channels near the inlet/outlet and the largest channel in the center of the plate. The land widths remain constant at 0.8 mm, but Zhang et al. do not discuss the reason for this result. CFD contour plots of the flow velocities are shown for the initial and optimal bipolar plate designs. The final design results in a much improved flow distribution with the velocity inside the channels being almost uniform.

Xing et al. [62] optimized the current density of an air-breathing fuel cell at a cell voltage of 0.7 V and 0.4 V. Two optimization problems are solved with one design variable each. The design variable is either: (a) the cathode open ratio or, (b) anode channel ratio. The optimization problem is solved using the SQP algorithm in MATLAB. With only one design variable, it is difficult to understand why an optimization algorithm was used to obtain the optimal design since a parametric study would have been sufficient. Numerical optimization is only advantageous as the number of design parameters increases because all parameters are modified at once.

Most recently Jang et al. [72] aimed to maximize the performance of a single cell with a 1 cm^2 active area. Their model introduces baffles in the channel, i.e. an obstruction is built into the bipolar plate in order to force air and hydrogen in the cathode and anode, respectively. The model is solved with CFD-ACE+ and the SCGM is used to optimize the position of the baffles in each channel for maximum performance. Following a similar optimization formulation to Cheng et al. [58], the objective function minimized by Jang et al. is the inverse of the current density rather than the negative. Nevertheless, optimal solutions were obtained for low and high flow rate cases in 4–5 days of computing time. The justification for the inclusion of the baffles is improved water removal from under the land area by convection and a "subtle" effect on the performance of the cell. The results presented show an increase in performance of 14% with respect to a base case with baffles midway along each channel, but make no comparison between a case with no baffles. Furthermore, the effects the baffles have on water vapour distribution and pressure drop within the cell are ignored. While this is an interesting application of shape optimization, the practical application of the optimization results should be further investigated.

3.1.2. Electrode optimization

Song et al. [63,64] pioneered the use of numerical optimization for electrode design. In their publications, they optimized the catalyst layer composition of a PEM fuel cell in order to achieve maximum current density at a specified voltage of 0.6 V. In both publications, a one-dimensional, macro-homogeneous catalyst layer model was used to analyze the cathode catalyst layer. The numerical model was validated with respect to experimental data in Ref. [73]. In Ref. [63], the design variables that are used are the Nafion volume fraction, the platinum loading, and the thickness of the catalyst layer. The optimization problem is solved with respect to only one or two of these design variables, therefore not taking full advantage of using numerical optimization. The MATLAB SQP solver *fmincon* is used to solve the optimization problem. The optimization results show an optimum ionomer loading of approximately 30 wt%, in agreement with previously reported experimental data [6,3].

In Ref. [64], a functionally graded electrode is optimized. The design objective is to maximize current density. The design variable is to obtain either the optimal Nafion volume fraction distribution or the platinum loading distribution in the thickness of the catalyst layer. In another case, both Nafion and platinum loading distributions are optimized simultaneously. The thickness of the catalyst layer is assumed to be 25 μm. The results show that the optimal distribution contains the most amount of Nafion and platinum near the membrane. Both Nafion and platinum loading decrease as the GDL is approached. In this study, the MATLAB SQP solver *fmincon* is also used to solve the optimization problem. The numerical optimization results are in agreement with experimental data in [74].

Rao and Rengaswamy [75] looked specifically at the optimization of one agglomerate in a CL. They presented two optimization studies: (a) minimizing the amount of platinum inside an agglomerate at a given voltage and current, and (b) maximizing the current generated in the agglomerate. A third formulation is also presented where the current densities at several points in the polarization curve are optimized simultaneously, leading to a multi-objective problem. The design variables are the platinum to carbon ratio in several radial shells of the agglomerate, i.e. the platinum to carbon ratio distribution within the agglomerate. No attempt was made to include the agglomerate model presented into a CL model. The governing equations of the agglomerate model are discretized using finite differences and are included in the optimization problem as constraints. The combined analysis-optimization problem is solved using the SQP algorithm in MATLAB *fmincon*.

Secanell et al. used a two-dimensional, through-the-channel, macro-homogeneous cathode electrode model [76,68] and a two-dimensional, through-the-channel, agglomerate cathode electrode model [69] to obtain the CL platinum loading, Nafion loading and platinum to carbon ratio, and GDL porosity that maximized the cell current density at a given operating voltage (0.6 V). To solve the optimization problem, a gradient-based interior point optimization algorithm was used in conjunction with analytical sensitivities (direct method). Since the problem is nonlinear, using the direct method to obtain the analytical sensitivities resulted in large computational savings and a good convergence to the optimal solution. Due to the low computational requirements of this method, an electrode model with a state-of-the-art catalyst layer model was used to predict fuel cell performance. In Ref. [69], the optimal composition of the electrode for different microstructures and under different operating conditions is presented. The results led to the conclusion that a large improvement in fuel cell performance could be achieved by increasing the weight ratio of platinum to catalyst support. This conclusion has been confirmed by the recent trend of fuel cell manufacturers to use 46% platinum weight ratio catalyst instead of the previously used 20% weight ratio catalyst. The optimal Nafion loading is predicted to be slightly higher than previous experimental studies; however, this is shown to be a result of the increased platinum to carbon ratio.

Secanell et al. performed the first attempt at trying to optimize an anode catalyst layer [70]. The anode design does not usually limit fuel cell performance; therefore, the objective was to minimize platinum loading. In order not to reduce the cell performance, the value of the anode current density was constrained to 1.5 A cm^{-2} at an overpotential of 15 mV. The design variables again were the CL platinum loading, Nafion loading and platinum to carbon ratio, and GDL porosity. Further, the anode catalyst layer thickness was also optimized. The results from this study showed that the optimal anode CL would have a thickness of 1 μm and a platinum loading of 0.0025 $mg_{Pt} \cdot cm^{-2}$. For this study, an in-house electrode model and an interior-point method were used for the analysis and optimization simultaneously.

Secanell et al. have recently presented an optimization formulation to design a complete MEA [77]. The MEA model is a two-dimensional, through-the-channel, isothermal, isobaric model. It includes the previously optimized agglomerate anode and cathode electrode models, and a proton conducting membrane. The design objective was to maximize current density. The design variables were the CL platinum loading, Nafion loading and platinum to carbon ratio, and GDL porosity in both anode and cathode. By optimizing the composition of the two electrodes in the MEA simultaneously, the design process accounted for the electrode composition and microstructure as well as their interaction. The article showed that the optimal compositions in the anode and the cathode are remarkably different.

Using the optimization framework presented in reference [77], Secanell et al. [44] optimized both performance and platinum loading. To illustrate the trade-offs between the two objectives, a Pareto set is presented. To solve the multi-objective problem, the weighted sum method was used. The method provides good results, and it is readily implemented in the optimization framework DAKOTA [78]. In this article, the first multi-objective optimization framework was presented to quantify the trade-offs between cost and performance in MEA design. Using this methodology, it was shown that a substantial increase in performance can only be achieved by increasing platinum loadings in the range of 0.1–0.5 mg cm^{-2}. For higher loadings, the increment in performance is marginal, and does not justify the increased cost.

Zhang et al. [53] optimized the porosity, permeability and thickness of the anode and cathode GDLs and the inlet gas stoichiometry in order to minimize the cathode overpotential at a cell voltage

of 0.6 V. They used CFD-ACE+ to solve a three-dimensional single cell model with seven serpentine channels. The optimization problem was obtained using Powell's algorithm, a non-gradient based method based instead on the conjugate directions algorithm. Performance of the optimization algorithm was not reported in the article. They minimize the potential drop in the cathode with the aim of creating a more active cathode catalyst layer to increase performance. The results show that at the design voltage of 0.6 V, the current density distribution within the cathode is more uniform. Interestingly, the total current produced by the cell does not change from the reference case at this voltage; the optimized design only seems to perform better at cell voltages below 0.5 V.

Jain et al. [79] extended the cathode agglomerate model presented by Secanell et al. [69] in order to obtain optimal platinum loading distributions in the CL at either a given cell voltage or current density. The optimal distribution of platinum shows the same trends as those reported by Song et al. [64], i.e. higher loading near the membrane. Jain et al. also solved the current density maximization problem in reference [69] using a nonlinear interior point method (gradient-based method) implemented in IPOPT. Excellent agreement was obtained. Following the approach of Rao and Rengaswamy [75], the model governing equations are discretized using a finite difference method and are included as constraints of the optimization problem. This approach, usually referred as all-in-one or simultaneous analysis and design approach (SAND), can substantially reduce computational costs because the optimization algorithm is used to solve the governing equations and the optimization problem simultaneously.

Basri et al. [80] have recently used numerical optimization to minimize the cost of a direct methanol fuel cell (DMFC). The article also presents an attempt at solving the numerical optimization process using a simultaneous analysis and design approach (SAND) [8,81]. In the SAND approach, the governing equations of the problem are included in the optimization algorithm as equality constraints. In Ref. [80], the equality constraints represent the governing equations of a zero-dimension model; however, the exact physical meaning of each governing equations is not given. The design variables in the optimization problem were not clearly defined since the authors did not differentiate between design parameters and values obtained in order to satisfy the governing equations. To solve the SAND optimization problem, the MATLAB routine *fmincon* is used where the initial design is given by solving the same problem using the GA library. As discussed previously, the authors of this review question the appropriateness of using a zero-dimensional model to obtain optimal platinum loadings and electrode thicknesses; therefore, the results from the optimization are not discussed. Even though the work in Ref. [80] uses a model that might be too simplistic for electrode design, this work is one of the first to use a SAND approach in fuel cell design. Using this approach could lead to great saving in computational time, especially if used in combination with multi-dimensional, high-fidelity fuel cell models.

3.1.3. Operating conditions optimization

Articles in the literature regarding optimization of the operating conditions can be divided in two categories: (a) optimization of operating conditions at the cell level [82,45,83–85]; and (b) optimization of operating conditions at the fuel cell system level [86,87]. This section discusses the former category. Fuel cell system optimization studies will be discussed in more detail in Section 4. It should be noted that the optimization of the operating conditions of the fuel cell level should involve both an accurate fuel cell model and a complete fuel cell system model. Otherwise, the optimization algorithm would always choose a high pressure and stoichiometry since these values are usually constrained by the power consumption of the air compressor [86].

Mawardi et al. [82] were the first to use numerical optimization for optimizing the operating conditions of a single cell. They used a one-dimensional, non-isothermal model to optimize the operating conditions of an MEA operating with hydrogen from a reformer. The objective function was to maximize power density at a given current density. The design variables were nine operating parameters, i.e., temperature, anode and cathode pressure, stoichiometry and relative humidity, nitrogen to oxygen mole fraction and carbon dioxide to hydrogen mole fraction. There are constraints for all design variables and three additional constraints: (a) minimum membrane hydration, (b) maximum temperature rise (in order to limit membrane degradation), and (c) maximum cell voltage (since size and capital cost increase at higher potential due to lower current densities). The Nelder–Mead simplex method combined with a simulated annealing algorithm was used to solve the optimization problem. These algorithms are non-gradient based methods and, therefore, the gradients are not necessary. Non-gradient based methods are usually more computationally expensive than gradient-based. In the paper, the number of calls to the analysis code and the computational time necessary to reach the solution are not discussed, so no comparisons can be made here. The optimization problem is solved for six different cases: (a) base case, (b) thinner membrane, (c) thinner electrode, (d) decreased min. cell potential, (e) increased max. temperature, and (f) CO in the anode stream. For each design, the optimization problem is solved at five different current densities. Results from the optimization show that the optimal operating conditions strongly depend on the current density and on the case being solved.

To the knowledge of the authors, Wu et al. [83] presented the only attempt in the literature at trying to account for the effects of system level performance when optimizing the operating conditions using a multi-dimensional fuel cell model. They also performed the first attempt at using a radial neural network as a surrogate model to reduce the computational time during optimization. Wu et al. [83] aimed at optimizing fuel cell system efficiency at low ($0.15\,A\,cm^{-2}$), medium ($0.45\,A\,cm^{-2}$) and high ($0.75\,A\,cm^{-2}$) current densities. The following objective is used to measure fuel cell system efficiency:

$$f(\mathbf{x}) = \eta_{sys} = \frac{AiV_{cell}}{\dot{G} + \dot{w}_{hum} + \dot{w}_{compressor}} \tag{6}$$

where A is the active area, i is the current density, V_{cell} is the predicted cell voltage, $\dot{G}$ is the maximum electrical power with the input fuel (accounting for stoichiometry and hydrogen recycling effects), $\dot{w}_{hum}$ is the humidifier power and $\dot{w}_{compressor}$ is the compressor power. The latter two values are obtained based on semi-empirical relations [88].

The design variables used were the cell temperature and the cathode gas pressure, stoichiometry and relative humidity. The multi-dimensional model was isothermal, single-phase, and consisted of independent models of different dimensionality for each layer that were then solved iteratively, e.g., a 3D model for the membrane and GDL, a 1D model for the cathode catalyst layer and a zero-thickness model for the anode catalyst layer. The multi-dimensional model was used to train a radial neural network that was then used for the optimization. A very limited discussion is given about how the radial neural network was trained. The optimization algorithm used was the feasible sequential quadratic programming (FSQP) method. Wu et al. [83] report optimal values for both an ideal system (no system losses) and a realistic system (including compressor and humidifier losses). For the ideal system, stoichiometry and pressure reached the upper bounds at medium and high current densities. For the realistic system, cathode stoichiometry was between 1.25 and 2 and pressure between 1.5 and 3 atm. The optimal cathode relative humidity was between 10% and

15% which seems extremely low. Based on the predicted efficiency at the optimal solution using the multi-dimensional model and the radial neural network, the neural network was able to predict the cell performance at the optimal point within an accuracy of ±2%. In this case, it seems that the neural network was trained only at the beginning of the optimization. It would be beneficial to train the neural network, or any surrogate model, during the optimization process in order to guarantee that the approximated optimization problem converges to the optimal solution of the high-fidelity model [89].

Chen et al. [45] investigated optimizing fuel cell operating conditions in order to minimize the fuel cell's capital and operating costs. The objective function was multi-objective with the following three objectives: (a) minimize the annualized cost of the cell, (b) minimize the fuel costs, and (c) maximize the credits for the exhaust hydrogen. The annualized cost of the cell was obtained as the ratio of cell active area to lifetime multiplied by a coefficient representing the cost of a square meter of active area. The second term was obtained by multiplyting the hydrogen flow rate by the cost of hydrogen. The latter term uses the same relationship but a different coefficient that represents the credit obtained by reusing the exhaust fuel. To predict the cell performance, a very simple zero-dimensional cell/stack model was used. There were two design variables: (a) the cathode overpotential, and (b) the hydrogen mole fraction at the outlet. All other operating conditions are obtained using the zero-order model, e.g. active area, anode pressure and cell voltage, The authors proposed two optimization algorithms: (a) a discretization method, and (b) a branch and bound interval analysis. The former is simply a parameteric study; therefore, in our opinion, it should not be considered as an optimization algorithm. Further, the only reason Chen et al. were able to use this method is because the number of design variables was two. The latter method is usually used for discrete design variables, therefore it might be very computationally expensive. Results from Chen et al. [45] suggested that electricity production rates with a fuel cell could be competitive with commercial rates; however, they also highlighted that the results have significant sensitivity to input parameters. Even though the fuel cell model is very simple and the number of design variables is small, Chen et al. work highlights that numerical optimization can be used, not only for optimizing performance, but also to minimize fuel cell costs, a critical issue for fuel cell commercialization.

Two articles have dealt with operating condition optimization in direct methanol fuel cells (DMFCs). Ko et al. [84] use a zero-dimensional, non-isothermal, mechanistic model to optimize the fuel efficiency of a DMFC. They first calibrate the model using a least-squares optimization method to determine kinetic parameters in the electrode. Anode and cathode flow rates and reactant concentrations are optimized over a time interval with varying load requirements using the dynamic optimizer DYNOPT. Wu et al. [85] extended this study and developed a multi-objective formulation. A similar model to Ref. [84] is used to maximize both the fuel efficiency and exergy efficiency, and minimize methanol crossover through the membrane. A fuzzy logic-based optimizer is employed to determine the optimal anode inlet temperature, bounded between 40 and 80 °C, at every time step for an interval with varying load requirements. The results for both studies show that there is much to be gained in the way of efficiency for dynamic control of DMFC systems without the need for extensive experimental testing.

3.1.4. Stack optimization

Two examples of stack optimization were found in the literature, and both are for PEMFCs. Wang and Dong [90] performed an optimization on their patented Tri-stream, External-manifolding, and Radiator Stack (TERS) using a zero-dimensional, isothermal, and semi-mechanistic stack model, along with an algorithm known as the Adaptive Response Surface Method (ARSM) developed in-house. The algorithm improves upon the Response Surface Method (RSM) by reducing the design space systematically to ensure the accuracy of the method approximation. The objective function is the system net power and the design variables are the fin height, fin thickness, and fin wavelength, all physical dimensions of the FC stack. The optimized dimension results compare favourably to a CAD model developed in Pro/Engineer where ANSYS is used to perform a stress/strain analysis. Mohamed and Jenkins [91] used a simple zero-dimensional, isothermal mechanistic stack model to find the optimal number of cells in series, as well as parallel, in the stack and the optimal cell membrane area to maximize power output. The optimization method is GA, and the mathematical modeling and optimization results are validated against empirical stack data. That there are only two optimization examples of PEMFC stacks shows that the area has yet to draw much in the way of research.

3.1.5. Summary

Table 1 shows a summary of optimization studies and their objective functions in fuel cell design. The design variables and constraints for theses studies are shown in Table 2. Table 3 shows the analysis and optimization codes used to solve the design problem.

3.2. High-temperature fuel cells

The number of articles dealing with numerical optimization of solid oxide fuel cells (SOFCs) is even more scarce than for PEMFCs. At the time of this review, the only attempt to use numerical optimization for SOFC single cell design was that of Bhattacharyya and Rengaswamy [92]. In their article, the geometry of a tubular SOFC is optimized in order to achieve two objectives: maximize

Table 1
Summary of fuel cell optimization studies: problem formulation.

Reference	Objective function
Channel design:	
Grujicic et al. [54]	max $i(V_{cell} = 0.7\,V)$
Grujicic et al. [55]	max $i(V_{cell} = 0.7\,V)$
Grujicic et al. [56]	max $i(V_{cell} = 0.7\,V)$
Lin et al. [57]	max $P(\phi_{cat} = 0.25\,V)$
Cheng et al. [58]	max $P(V_{cell} = 0.7\,V)$
Huang and Lin [59]	max $i(V_{cell} = 0.3\,V)$
Huang et al. [60]	max $i(V_{cell} = 0.7\,V$ or $0.4\,V)$
Wang et al. [61]	max $P(V_{cell} = 0.4\,V)$
Xing et al. [62]	max $i(V_{cell} = 0.7$ or $0.4\,V)$
Zhang et al. [71]	min equation (5)
Jang et al. [72]	max $i(V_{cell} = 0.7)$
Electrode design:	
Song et al. [63]	max $i(V_{cell} = 0.6\,V)$
Song et al. [64]	max $i(V_{cell} = 0.6\,V)$
Song et al. [64]	min $m_{Pt,agg}$
Rao et al. [75]	max $i_{agg}(V_{cell} = 1.05 - 0.4)$
Rao et al. [75]	max $\sum_{j=1}^{12} i_{agg}(V_{j,cell} = 1.04 - 0.45)$
Secanell et al. [68]	max $i(\phi_{cat} = 0.3\,V)$
Secanell et al. [69]	max $i(\phi_{cat} = 0.3, 0.5, 0.7\,V)$
Secanell et al. [70]	min $m_{Pt,a}$
Secanell et al. [77]	max $i(\phi_{cat} = 0.3, 0.5, 0.7\,V)$
Secanell et al. [44]	max $i(\phi_{cat} = 0.3, 0.5, 0.7\,V)$ and min m_{Pt}
Zhang et al. [53]	min $\eta(0.6\,V)$
Zhang et al. [53]	min m_{Pt}
Jain et al. [79]	max $i(\phi_{cat} = 0.3\,V)$
Operating conditions:	
Mawardi et al. [82]	max P_d (W/cm^2)
Wu et al. [83]	max η_{sys} ($i = 0.15$ or 0.45 or $0.75\,A\,cm^{-2}$)
Chen et al. [45]	min annualized cost
Ko et al. [84]	max η_{fuel}
Wu et al. [85]	max η_{fuel}, η_{sys} and min r_{Xover}

Table 2
Summary of fuel cell optimization studies: design variables and constraints.

Reference	Design variable	Constraints
Channel design:		
Grujicic et al. [54]	p_c^{in},$t_{GDL,c}$,w_c,$(w_{cc}/(w_{ch}+w_{cc}))$	Bounds in variables
Grujicic et al. [55]	$t_{GDL,c}$,$h_{ch,c}$,$w_{ch,c}$	bounds in variables
Grujicic et al. [56]	$t_{GDL,c}$,w_c,$(w_{cc}/(w_{ch}+w_{cc}))$	Bounds in variables
Lin et al. [57]	ϵ_V^{CL},ϵ_V^{GDL},w_{ch}/w_{cc}	None
Cheng et al. [58]	λ,h_{ch},t_{GDL}	None
Huang and Lin [59]	Height of the gas channel (3 DV)	None
Huang et al. [60]	Height of the gas channel (4 DV)	None
Wang et al. [61]	Height and width of the gas channel (8 DV)	None
Xing et al. [62]	Case 1: λ_c	None
Xing et al. [62]	Case 2: λ_a	
Zhang et al. [71]	Width of gas channel and land	Bounds in variables
Jang et al. [72]	Location of baffles in channel	Bounds in variables
Electrode design:		
Song et al. [63]	ϵ_N, $m_P t$, $t_{CL,c}$	$\epsilon_N+\epsilon_V+\epsilon_S=1$
Song et al. [64]	Either ϵ_N or $m_P t$ distribution	$\epsilon_N+\epsilon_V+\epsilon_S=1$
Rao et al. [75]	$Pt\|C$ (9DV)	Bounds in variables
Secanell et al. [68]	ϵ_V^{GDL},ϵ_{agg},$m_P t$,$Pt\|C$	Bounds in variables
Secanell et al. [69]	ϵ_V^{GDL},ϵ_{agg},$m_P t$,$Pt\|C$	Bounds in variables
Secanell et al. [70]	ϵ_V^{GDL},ϵ_{agg},m_{Pt},$Pt\|C$	Bounds in variables, $i(\phi_a=15mV)=const$
Secanell et al. [44,77]	$\epsilon_N^{CL,a}$, $\epsilon_V^{CL,a}$, $m_{Pt,a}$, $\epsilon_V^{GDL,a}$, $\epsilon_N^{CL,c}$, $\epsilon_V^{CL,c}$, $m_{Pt,c}$, ϵGDL, c_V	Bounds in variables
Zhang et al. [53]	$\epsilon_V^{GDL,a}$, $\epsilon_V^{CL,a}$, $t_{GDL,a}$, $k_{GDL,a}$, λ_a, $\epsilon_V^{GDL,c}$, $\epsilon_V^{CL,c}$, $t_{GDL,c}$, $k_{GDL,c}$, λ_c	Bounds in variables
Jain et al. [79]	ϵ_V^{GDL},ϵ_{agg},$m_P t$,$Pt\|C$	1: Bounds in variables 2: Bounds in variables 3: Bounds in variables
Operating conditions:		
Mawardi et al. [82]	T_{cell}, p_c, p_a, RH_c, RH_a, x_i, (9DV)	Bounds in variables,λ_N, V_{max}
Wu et al. [83]	T_cell, p_c, λ_c, RH_c	Bounds in variables
Chen et al. [45]	Overpotential,H_2 Mole fraction out	Bounds in variables
Ko et al. [84]	$\dot{V}_c$,$\dot{V}_a$,λ_c,λ_a	Bounds in variables
Wu et al. [85]	T_a^{in}	Bounds in variables

Table 3
Summary of fuel cell optimization studies: analysis and optimization codes.

Reference	Analysis code	Optimization code
Channel design:		
Grujicic et al. [54]	COMSOL	MATLAB—*fmincon*
Grujicic et al. [55]	COMSOL	MATLAB—*fmincon*
Grujicic et al. [56]	COMSOL	MATLAB—*fmincon*
Lin et al. [57]	In-house	*SCGM*
Cheng et al. [58]	CFD-ACE+	*SCGM*
Huang and Lin [59]	CFD-ACE+	*Maquardt*
Huang et al. [60]	CFD-ACE+	*Maquardt*
Wang et al. [61]	FLUENT	*SCGM*
Xing et al. [62]	COMSOL	MATLAB—*fmincon*
Jang et al. [72]	CFD-ACE+	*SCGM*
Electrode design:		
Song et al. [63]	MATLAB 1D CL model	MATLAB—*fmincon*
Song et al. [64]	MATLAB 1D CL model	MATLAB—*fmincon*
Rao et al [75]	Maple-MATLAB	MATLAB—*fmincon*
Secanell et al. [68]	In-house	DOT—*SQP*
Secanell et al. [69]	In-house	DAKOTA—*OPT ++*
Secanell et al. [70]	In-house	DAKOTA—*OPT ++*
Secanell et al. [77,44]	In-house	DAKOTA—*OPT ++*
Zhang et al. [53]	CFD-ACE+	*Powell*
Jain et al. [79]	In-House-AMPL	IPOPT
Operating conditions:		
Mawardi et al. [82]	In-house	*Nelder-Mead*
Wu et al. [83]	In-house	FSQP
Chen et al. [45]	In-house	Branch-and-bound
Ko et al. [84]	In-house	DYNOPT - *SQP*
Wu et al. [85]	In-house	*Fuzzy set multi-objective*

the gravimetric power density in order to reduce cost, and maximize the volumetric current density in order to reduce cell size. The design variables are: (a) radius of the anode channel, (b) the cell length, and (c) the annulus size. The multi-objective optimization problem is formulated using a so-called lexicographic approach. Using this approach, a first optimization problem is solved that has only the first objective function, deemed the most important to the designer. The second objective problem is then solved separately using an additional constraint in order to guarantee that the value of the original objective is equal or larger than the value obtained in the first optimization. If there are more than two objectives this process is continued. Bhattacharyya and Rengaswamy are the first to apply this approach to solve a multi-objective fuel cell design problem. In order to predict the performance of the fuel cell, a two-dimensional (radial and along the tube), isothermal, non-isobaric SOFC model is used. A zero-thickness model is used to model the catalyst layers. The model is validated against experimental data at two temperatures and two hydrogen flow rates. To solve the system of PDEs resulting from the governing equations, the PDEs are discretized in MAPLE by forward/backward differences. Then, the resulting nonlinear system is exported to MATLAB and solved using the nonlinear solver *fsolve*. The multi-objective single cell design problem is solved using the SQP function *fmincon*. The optimization problem is solved at different voltages and at two operating conditions: $T=800$ K, 45 ml/min and $T=850$ K, 35 ml/min. The latter optimal design achieved 30% and 65% increases in gravimetric and volumetric power densities, respectively. The changes in the radius of the anode channel, cell length, annulus size were 22%, 8%

and 20% respectively. These changes remained very similar at any cell voltage from 0.9 V to 0.5 V.

The development of computer software that is capable of providing analytical sensitivities is critical in order to be able to perform numerical optimization using multi-dimensional codes with a reasonable amount of computational resources. The works of Kapadia et al. and Elliott et al. are paving the way towards introducing multi-dimensional numerical optimization to SOFC design [65,66,93]. Even though they have not yet performed any numerical optimization studies, these references introduced a multi-dimensional model for SOFCs that was capable not only of predicting the fuel cell performance but also of obtaining the sensitivities of several functionals, such as current density, with respect to different design parameters such as anode and cathode porosity. Sensitivities were obtained using forward-differences as well as analytical differences using both direct and adjoint methods.

4. Fuel cell systems integration

The objective of the optimization for fuel cell systems (FCSs) is usually either the improvement of a given design by varying system operation parameters or the improvement of the design by varying system physical characteristics while keeping the operating parameters constant. In rare cases, a combination of these two objectives has been realized in the literature. Optimizations of system operation parameters are much more common for fuel cell systems optimization than it is for single fuel cell optimization where cell design is more prevalent. This section is a review of the literature for FCS optimization, but is limited in scope to stand-alone FCSs and does not consider hybrid systems.

FCSs can be categorized initially by the type of fuel cell: PEMFC and high-temperature fuel cell systems are the two main types that have been optimized in the literature. There are many more optimization studies of PEMFC-based systems than high-temperature systems. The next categorization is by optimization objective, either changing certain aspects of the design to arrive at an optimal design or improving the operation of a given FCS design. The third categorization is by model type: theoretical (mechanistic), semi-empirical, and empirical, as discussed in Cheddie et al. [94]. From an examination of the literature, zero-dimensional, steady-state, isothermal, semi-empirical models of PEMFC-based systems are most commonly used to optimize the operating parameters. From the authors' perspective, the full value of optimization has not yet been realized for the optimization of FCSs, where there is a conspicuous absence of a validated transient, three-dimensional, theoretical model with optimized physico-chemical parameters. It must be noted that such a model is rare in the literature and the level of complexity would likely make the computational resources required to solve the optimization problem onerous. However, these deficiencies illustrate that the field of FCS optimization is relatively immature and there is substantial room for more ambitious optimization attempts.

The following two sections will include a review of optimization studies in PEMFCs and high-temperature fuel cell systems found in the literature. The categorizations from above will be used to group similar attempts and to distinguish between the various optimizations.

4.1. Low-temperature fuel cell system optimization

The review of the optimization of PEMFC systems will first include the operating parameter optimizations, followed by non-numerical system design optimizations, and finally the attempts at physico-chemical optimizations.

4.1.1. Operating parameters optimization

Five research groups used zero-dimensional, steady-state, semi-empirical models for operating parameter optimizations. Blunier and Miraoui [95] optimized the net system voltage at a given current by varying the system pressure and the air stoichiometry. The objective of the study is to elucidate the effect of air humidity in the stack outlet. It was concluded that fully humidified air at the stack inlet can be detrimental to voltage at low airflow rates because high air stoichiometry is required to avoid flooding of the electrode. At high airflow rates, full humidification is required to avoid membrane drying. Wishart et al. [86,96] used the stack model of Mann et al. [97] to perform single- and multi-objective optimizations of net system power and efficiency by varying stack pressure, temperature, and air stoichiometry. Local (SQP) and global (simulated annealing and genetic algorithms) optimization algorithms were compared for efficacy and solution speed. The multi-objective optimization results were applied to a low-speed hybrid electric vehicle undergoing the New York City Cycle (NYCC) drive cycle. Na and Gou [98] optimized the system efficiency and cost with respect to system pressure, reactant stoichiometric ratios, stack voltage, and stack current as the design variables. A sequential quadratic programming (SQP) was used to solve the problem; therefore, it is unclear whether only local optimal solutions were found. Xu et al. [99] used the commercial Aspen Plus software to model the reformer of a PEMFC system and a simple theoretical, steady-state, zero-dimensional stack model from Godat and Marechal [100]. The design variables were the steam methane reformer (SMR) temperature, water gas shift (WGS) temperature, and feed rate of methane. These design variables were determined from a sensitivity analysis study performed in Aspen Plus. Using water instead of steam as the process feed was also considered. The objective functions are efficiency and cost and the optimization algorithm was SQP. The results show that there is a benefit to performing the cost and efficiency optimizations simultaneously rather than sequentially. Lastly, Xuan et al. [101] use a response surface method (RSM) to optimize a PEMFC stack model while considering balance of plant (BOP), including an air pump and cooling fan in their formulation of stack power. The optimization problem is multi-objective, minimizing fuel consumption while maximizing stack power using the stack current, temperature, and stoichiometry and relative humidity of the input streams as design variables. The conclusion from all these optimization attempts is that judicious selection of operating parameters has a significant impact on FCS operation, and that this type of optimization should occur for any design.

4.1.2. Integer system design optimization

Two research groups have incorporated non-numerical system design options in the optimization, where the binary variables result in mixed-integer, non-linear programming (MINLP) problems. Marechal et al. [100] solved a MINLP problem using process integration. The optimization incorporated an evaluation of a steam methane reformer (SMR) and partial oxidation and reforming reactor (POX) for hydrogen production and an evaluation of a water gas shift (WGS) reaction using either a medium-temperature reactor or two-step (high- and low-temperature) reactor system for post-reforming CH_4 processing. The stack model is zero-dimensional, steady-state and mechanistic. The design variables are the steam to carbon ratio, oxygen to nitrogen ratio, oxygen to carbon ratio, fuel processing temperature, fuel utilization, and post-combustion pressure. The optimization uses genetic algorithms, and it is thermo-economic, thereby maximizing system efficiency and minimizing system cost. Kamarudin et al. [102] employed a methanol feed and reformer action to obtain hydrogen for a PEMFC system. Five reforming methods are used as design variables, as are reactant flow rates and molar fractions. The resulting MINLP is solved using the SQP algorithm. The five alternatives are optimized for

CO output as well as investment and manufacturing costs. The FC stack is the zero-dimensional, steady-state, semi-empirical model of Mann et al. [97]. The objective of the study was to apply the superstructure-based approach to the optimization of the reactor-separation network of the FCS.

Most recently, Han et al. [103] presented a model of a fuel cell vehicle with an integrated quasi-static model of a fuel cell stack. They validate their model against a fully dynamic fuel cell model by simulating loading of three typical drive cycles and achieve agreement to within 5% of overall fuel efficiency with a 98% reduction in simulation time. The vehicle model considers driving dynamics, powertrain performance and weight of all critical components. The goal of the optimization is to maximize the fuel economy of the vehicle with respect to several integer design parameters, such as number of cells in the stack, number of battery units, and continuous design variables, such as gear ratios and power control limits. All the variables are bounded, and further design constraints are imposed related to performance such as acceleration and top speed. The algorithm also contains an inner optimization loop which maximizes the stack power at every iteration by selecting the optimal air excess ratio, subject to flow rate constraints corresponding to the operating limits of the compressor. Two different algorithms, DIRECT and NOMADm, optimize the system with an increase of 14% in fuel efficiency from the base case, but with a slightly different set of design variables. Han et al. [103] extend the study and use NOMADm to optimize the system when regenerative braking is included in the model. Here, an 18% increase in fuel efficiency is observed. This type of model and optimization can be expanded to full vehicle design with additional space (packaging) and cost constraints.

4.1.3. Physico-chemical optimization

Finally, there are three examples of physico-chemical optimizations in the literature. Xue and Dong [104] used a semi-empirical, steady-state model of a Ballard Mark IV fuel cell stack from Mann et al. [97] and models for auxiliary systems such as the air compressor in order to create a comprehensive fuel cell system model. Using this model and joint concurrent optimization, the optimal active stack area and air stoichiometry ratio were obtained to maximize net power output, and, at the same time, minimize production costs.

Wang and Dong [105] optimized a fuel cell system as a demonstration of a novel optimization algorithm called the ARSM. The stack model was again the model from Mann et al. [97]. The design variables were the air stoichiometry, stack width, supporting column width, number of cells in the stack, end plate thickness, manifold cover thickness, and the height of the fins in the stack. A sensitivity analysis showed that the air stoichiometry, end plate thickness, manifold cover thickness, and fin height did not have a large impact on the net power, and so only the stack width, supporting column width, and number of cells were used in the optimization. The optimization results showed an improvement in system efficiency, net power, volumetric and gravimetric power densities, as well as in a reduction of system costs.

Ang et al. [106] used the system efficiency and MEA area as design variables to determine the optimal values for a given net system power. The fuel cell model is for a single-cell stack, and the model is isothermal and isobaric as well as zero-dimensional. The optimization algorithm is LINDOGlobal, a commercial solver available from Lindo Systems, Inc. The algorithm is a branch-and-cut method that disaggregates an NLP into smaller problems. Ang et al. [106] conclude that for a given power output that a more efficient system has a larger MEA and vice versa. The multi-objective optimization reveals that system efficiency should remain between 40% and 47% and that the MEA size should be at least as large as $3\,cm^2\,W^{-1}$.

4.1.4. Summary

The optimization examples of PEMFC systems highlight the need for more ambitious optimization attempts. None of the models are transient and all are zero-dimensional. As confidence in stack models increases, the computational resources necessary for solving stack models are reduced and optimization algorithms are evaluated, it is expected that the level of fuel cell system complexity used will increase. Table 4 lists a summary of the PEMFC-based system optimizations discussed in this section.

4.2. High-temperature fuel cell system optimization

High- and medium-temperature FCS optimization has been attempted by several research groups but the list of examples is not extensive. If interest in this type of fuel cell continues to grow, more widespread modeling and optimization efforts can be expected.

Two research groups have explored the optimization of molten carbonate fuel cell (MCFC) systems. Yong et al. [107] make use of a zero-dimensional, steady-state model for a molten carbonate fuel cell (MCFC) system, where fuel consumption is minimized by varying the reactant flow rate and cathode gas recycle ratio. The optimization is performed using genetic algorithms. The optimization results are validated using an experimental 10 kW MCFC system designed for stationary, residential applications. Chudej et al. [108] optimize the control strategy of a MCFC in order to balance the allowed speed of load changes and the allowed temperature differences inside the cell. The objective is to ensure

Table 4
Summary of fuel cell system optimization studies: problem formulation and optimization algorithm.

Reference	Objective function	Optimization algorithm
Operating conditions:		
Blunier and Miraoui [95]	Inlet air pressure and stoichiometry	Not given
Wishart et al. [86,96]	System power and efficiency	SQP Genetic algorithms Simulated annealing
Na and Gou [98]	System efficiency and cost	SQP
Xu et al. [99]	System efficiency and cost	SQP
Integer:		
Marechal et al. [100]	System efficiency and cost	In-house
Kamarudin et al. [102]	Cost and CO production	SQP
Han et al. [103]	Fuel efficiency	MATLAB—*NOMADm*
Physico-chemical:		
Xue and Dong [104]	System power, efficiency, and cost	Not given
Wang and Dong [105]	System power and cost	Adaptive response surface
Ang et al. [106]	System efficiency and MEA size	LINDOGlobal

Table 5
Summary of high- and medium-temperature fuel cell system optimization. Problem formulation and optimization algorithm.

Reference	Objective function	Optimization algorithm
MCFC:		
Yong et al. [107]	Fuel consumption	Multi-crossover genetic algorithm
Chudej et al. [108]	Temperature time gradients	Sparse SQP (SNOPT)
SOFC:		
Barratto and Diwekar[109]	System efficiency, cost, and environmental impact	MINSOOP
Palazzi et al. [110]	System efficiency and cost	In-house
Vijay et al. [111]	System efficiency	MATLAB—*fminsearch*
PAFC:		
Zervas et al. [112]	Fuel consumption	GAMS

that the material's thermal stress resulting from large temperature gradients does not compromise the structural integrity. The optimization is performed on a complex 2D crossflow model and results in a large number of partial differential equations (PDEs) as part of a Pareto-optimal control problem. The results are validated against a closely related model of an actual MCFC system, and the authors conclude that as the system models grow in complexity, model reduction techniques such as proper orthogonal decomposition may become necessary.

Other research groups attempted optimization of SOFC-based systems. Barratto and Diwekar [109] performed a multi-objective optimization of efficiency, environmental impact, cost, and health impact using the SOFC model in the Aspen Plus process simulation to be used in auxiliary power units (APUs) of heavy-duty vehicles at rest stops in the South California Air Basin. The design variables were diesel fuel intake, reformer temperature, system pressure, cathode air stoichiometric ratio, air pre-heater temperature, and fuel utilization in the stack. The optimization resulted in a set of alternative designs that optimize cost, efficiency, and environmental and health impacts separately: no relative weighting of these objectives was assigned. Palazzi et al. [110] used the BELSIM-VALI and EASY software developed in-house to conduct a thermo-economic optimization of the design of a 50 kW, planar SOFC system for stationary applications. The optimization of minimizing cost and maximizing efficiency uses the Pinch method and genetic algorithm to produce the Pareto curves that demonstrate the trade-offs of the two objectives, and can be used in making design decisions. Vijay et al. [111] use an SOFC system model with inlet stream heat exchangers and an afterburner and aim to maximize the efficiency. MATLAB's *fminsearch* is used for the optimization with the anode and cathode flow rates as the design variables. They investigate the trade-offs between operating the cell at maximum efficiency or at constant fuel utilization across its operating range. The results show that the tradeoffs between the two cases are small.

A single medium-temperature fuel cell system optimization could be sourced in the literature. Zervas et al. [112] employ a fully three-dimensional, CFD, steady-state, isothermal model in a meta-model approach that generates a database of system variables. Linear regression and non-linear neural-network models (NNMs) are both used to develop the correlation between inputs and outputs and in the optimization of the NLP problem. The CFD software is PHOENICS, and the SIMPLEST algorithm is used to solve the resulting PDEs. The optimization objective is to reduce first the fuel volumetric flow rate and second the oxidant volumetric flow rate and the different modeling and optimization methods used demonstrate that various approaches can be used in this type of optimization.

4.2.1. Summary

The number of optimization examples of high-temperature FCSs is quite low, and this indicates that this area of research has not been fully explored yet. As in the case of PEMFCS optimization attempts, more detailed models are required in order to substantially increase the performance of high-temperature FCSs. As the industry matures, the number of modeling groups is expected to similarly increase. Table 5 lists a summary of the high- and medium-temperature FCS optimizations. As in the previous sections, the constraints of the optimization were found to all be bounds in the variables.

5. Conclusions

Fuel cell optimization is a relatively new area of research. Only in the last decade have articles in the area of fuel cell optimization started to appear in the literature. Competition to provide the most cost effective, efficient and durable fuel cell, and advancements in fuel cell computational models will increase the interest in numerical optimization of fuel cells. Based on the current literature survey, the state of the art in fuel cell design and optimization has already been able to provide insight that would have been difficult to obtain by trial-and-error. New optimization formulations that include multiple objectives, e.g. cost, reliability and durability, and additional design variables, e.g. the shape and topology of the fuel cell flow channel, can provide further insight into the optimal fuel cell geometry and composition and will lead to innovative designs that cannot be designed by trial-and-error such as variable cross-sectional area flow channel bipolar plates.

In the area of channel design, previous optimization studies have mainly focused on optimizing current density. The design variables have been the channel to land ratio and the channel dimensions. The related assumption has been that the shape of the channel would remain rectangular and that the cross-section would be uniform. Only in the past two years, several research studies have started to aim at predicting the optimal shape for channels with varying geometry. Unfortunately, these studies have been limited to a small number of design parameters and to small sections in the gas channel. Therefore, research remains to be done on finding a formulation to optimize the shape of the channel along the flow direction. Channel design studies have also ignored the tradeoffs between increasing current density and decreasing pressure drop. Future work is also necessary to develop a multi-objective formulation that accounts for both of these objectives simultaneously.

All previous optimization studies in channel design have been concerned with the shape of the channel. Obtaining the optimal topology of the channels – e.g, serpentine, straight – might provide substantially larger improvements in performance. For example, a recent experimental study achieved an improvement of up to 30% in fuel cell performance by changing the topology of the channels in the flow field [113]. Topology optimization [13] can be used as an innovative method to obtain the optimal layout and geometry of the fuel cell flow field. Topology optimization for fluid flow has only recently been attempted [114].

In the area of electrode design, previous work has used average porosity, platinum and Nafion loadings as design variables. These values dictate the structure of the catalyst layer. Future work must include the development of relationships between fabrication parameters and the final catalyst layer structure. Most articles have been concerned with maximizing the current density of the electrode. Cost, reliability and durability have now become the main barriers to fuel cell commercialization. Multi-objective formulations such as the ones developed by Chen et al. [45], Rao and Rengaswamy [75] and Secanell et al. [44] must continue to be developed. Since electrodes are three-dimensional structures, future work is also necessary in order to obtain functionally-graded electrodes that can adapt to the changing concentration of the reactants in the channels. To achieve this goal, the number of design variables used for designing the electrode must be increased substantially and a three-dimensional electrode model will be necessary.

In order to achieve realistic optimal designs, it is also necessary to increase the accuracy of the fuel cell models and to analyze the range of applicability of the models. Most of the articles in channel and electrode optimization use steady-state models. Models for channel optimization usually rely on either a zero-thickness or a macro-homogeneous electrode model. Electrode optimization studies rely on either a macro-homogeneous or an agglomerate model for predicting the CL performance. Recent models that include the pore size distribution should be used in the future. Further, in order to optimize durability, transient models will be necessary.

The current state of the art for FCS optimization is similar in some respects to that for the cell level. For the most part, the models that have been used have been steady-state and isothermal, without the required complexity to be both accurate representations of real-world systems and flexible enough to be applicable to a wide range of designs. As the models at the cell level increase in complexity and accuracy, the system models that incorporate the former will follow suit. However, much work needs to be done in the modeling and optimization of the subcomponents of the system such as the air compressor. In most of the optimizations, the design variables are the operating parameters; while improving the operation of a given FCS is useful, the true promise in using optimization in FCSs is to improve the design and manufacture processes, of which Xue and Dong [104] is an early, simple example.

Experimental validation of numerical optimized designs has only been performed in one study [59]. In order to increase the confidence in numerical optimization, a thorough experimental validation of the optimal designs obtained by numerical simulation is necessary. The benefits of validating optimal designs go beyond making sure that the design outperforms previous designs. Developing a new design based on optimization and testing its performance is the best method to test the predictive capabilities of current analysis codes. Further, by analyzing the parameters for which the optimal designs are not properly predicted, the physical phenomena that is poorly understood can easily be highlighted and can lead to development of new fuel cell mathematical models.

There are also other research areas that can benefit from improved numerical optimization methodologies. Some of the key areas that can be improved using optimization algorithms are the areas of parameter estimation and uncertainty analysis. Several parameter estimation studies have recently appeared in the literature [115,116,67,117]. However, a parameter estimation technique coupled with a multi-dimensional fuel cell model containing an accurate catalyst layer model still needs to be developed. Such a tool would be extremely important to identify the value of some of the key parameters in current fuel cell catalyst layer and single cell models such as agglomerate size and ionomer coverage. Further, to date, parameter estimation has only been used to fit polarization curves; however, segmented cells offer the opportunity to fit the current distribution in a single cell. This information could be used to obtain the key input parameters for multi-dimensional fuel cell models.

Last but not least, in order to solve large-scale optimization problems with tens or thousands of design variables, fuel cell analysis models that include analytical sensitivity analysis need to be developed such as the models of Kapadia et al. [66] and Secanell et al. [77].

In light of the recent progress made in new materials for fuel cells, the necessity for design methodologies that are fast and reliable is tremendous. The potential of novel materials can only be harvested by developing designs and operating conditions that maximizing the advantages of the materials while minimizing their weaknesses. If a novel material is introduced in a current cell design, it might only perform slightly better or even worse simply because the design was optimized to achieve the best performance for the current materials. Given the amount of proposed new materials for fuel cells (e.g., new short-side chain PFSI and hydrocarbon membranes, core-shell catalysts and new catalyst supports) and the extensive time and money requirements of the trial-and-error design approach, computational design and optimization is critical to the development of new fuel cell designs that can take full advantage of these new materials.

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