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Design Optimization and Parameter Estimation of a PEMFC using Nature Inspired Algorithms

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Abstract Numerical simulation of a Proton-Exchange Membrane Fuel Cell (PEMFC) requires an adequate model and precise parameters for reproducing their operational performance quantified by the polarization curve. Bio-inspired algorithms are well-suited for estimating unknown parameters; nevertheless, the simulator is stressed by inputting thousands of randomly-generated parameters. As a consequence, they require a robust numerical model. Once the model with the proper parameters reproduces the experimental data, it is applicable for optimizing the PEMFC. This article proposes a reformulation of a macro-homogeneous mathematical model by analyzing its relation with singularly perturbed differential equations to provide higher numerical stability to the solutions. This formulation introduces a boundary value problem instead of the usual initial value problem. Hence, we propose an assembly of the proper numerical methods for solving this version of the problem. Then, we introduce optimization problems for parameter estimation and design optimization; three bio-inspired algorithms solve the first. The most consistent of them is applied to optimize the PEMFC design by maximizing its performance or minimizing the platinum mass loading, highly related to the cost. To the best of our knowledge, there is no other research for optimizing the cell design, neither its performance nor its cost, using numerical simulation and optimization. The results are validated as follows: a) Comparing experimental polarization curves with the simulation with the parameters estimated by bio-inspired algorithms. b) Comparing a base design's performance with the optimized for maximum performance, and c) comparing a base design with the optimized for minimum platinum mass loading.

Keywords PEM Fuel Cell · Optimization Algorithm · Evolutionary Algorithms · Parameter Estimation · Macro-homogeneous mathematical model · Minimization

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of platinum mass loading · Performance maximization · Genetic Algorithm · Particle Swarm Optimization · Univariate Marginal Distribution Algorithm with Gaussian Model

1 Introduction

Fuel cells are devices of great interest since they have high efficiency converting hydrogen and oxygen's chemical energy directly into electricity by electrocatalytic reactions. In recent decades, great research efforts have been made for its development at a commercial level. There are various applications of interest, such as stationary power generation, trains, buses, cars, or other transports, including airplanes, trying to achieve a hydrogen-based economy [1].

A Proton Exchange Membrane Fuel Cell (PEMFC) is an electrochemical device that consists of an ionomer membrane in contact on either side with a catalytic layer, a diffusion layer, and a terminal plate with flow channels for the reactants. Figure 1 shows the basic structure of a PEMFC. At the anode, hydrogen flows into the channels through the diffusion layer (DL) to the catalytic layer (CL), usually conformed by Pt/C catalyst. At Pt catalytic active sites, H_2 is oxidized to produce protons and electrons. Protons move to the cathodic catalytic layer through the membrane, while electrons move to the cathodic current collector via the external electric circuit. Meanwhile, oxygen flowing in the cathodic flow channels diffuses through the DL to the cathodic CL, usually based on Pt/C or PtRu/C catalysts. It is reduced with the electrons and protons to produce water and heat. These reactions are represented in Equations (1) to (3), [2,3].

Fig. 1 Basic structure of a proton exchange membrane fuel cell.

Anode reaction:

Cathode reaction:

Global reaction:

Table 1 Phase composition and governing equations parameters.

ϵ_c	Void volume fraction, or CL porosity.
$L_{m,c}$	Volume fraction of the ionomer phase.
L_s	Volume fraction of solid portion of the GDL.
ϵ_g	GDL porosity.
V_{tot}	Total CL volume.
l_c	Thickness of the CL (μm).
ρ_{Pt}	Platinum density ($kg\ m^{-3}$).
ρ_C	Carbon density ($kg\ m^{-3}$).
m_{Pt}	Mass loading of platinum per unit area of the cathode, $mg\ cm^2$.
m_C	Mass loading of carbon per unit area of the cathode, $mg\ cm^{-2}$.
f	Platinum mass fraction in the Pt/C catalyst.
$D_{O_2-m}^{eff}$	Oxygen effective diffusion coefficient in the membrane ($m^2\ s^{-1}$).
$D_{O_2-w}^{eff}$	Oxygen effective diffusion coefficient in the water-flooded void region ($m^2\ s^{-1}$).
D_{O_2-m}	Oxygen diffusion coefficient in the membrane ($m^2\ s^{-1}$).
O_2	Oxygen concentration ($mol\ m^{-3}$).
F	Constant of Faraday ($96485\ C\ mol^{-1}$).
i	Proton current density.
S	Stoichiometric coefficient.
n	Number of electrons exchanged.
i_0	Exchange current density.
α_c	Cathodic electron transfer coefficient.
α_a	Anodic electron transfer coefficient.
a	Specific area
A_s	Reaction surface area per unit mass of platinum ($m^2\ kg^{-1}$).
$i_{0,ref}$	Reference exchange current density.
$C_{O_2,ref}$	Reference oxygen concentration ($mol\ m^{-3}$).
γ_{O_2}	Order of the oxygen reduction reaction.
η_{act}	Overpotential of activation.
κ^{eff}	Effective protonic conductivity.
κ	Bulk protonic conductivity.
σ^{eff}	Effective solid electronic conductivity.
σ	Solid electronic conductivity.
R_{Ohmic}	Specific ohmic resistance.
E_{rev}	Reversible cell potential.

The electrode-reactions rate depends on the CL structural parameters such as composition, distribution of catalytic materials, and the manufacturing process. Mathematical models are important to provide useful information for studying a PEMFC performance and improving its design. Different studies related to modeling and simulation of a PEMFC were carried out by other authors [2,4,5,6,7,8]. However, mathematical models require a set of parameters that must be estimated because they are very difficult to measure experimentally.

In the literature, three principal class of mathematical modeling for the cathode catalyst layer have been studied: interface models [9,10,11], macro-homogeneous models [12,7,13,6,5,8], and agglomerate models[4,14,15]. The macro-homogeneous models are intermediate complexity and have the advantage of providing expected results at low and medium current densities [6]. This model stands out for being an intermediate alternative between the interface model and the agglomerated model, providing adequate information to study the relationships between the catalyst layer's physical parameters and the performance of the PEMFC [4,6]. Besides, they permit analyzing the performance of the fuel cell through the polarization curve, by using the transport and diffusion of species through the catalytic layer. A macro-homogeneous model consists of a set of coupled nonlinear differential equations that describe oxygen diffusion, overpotential, and current density [6,5].

In mathematical modeling of PEMFC, often, the proposals privileged simple and intuitive governing equations to implement numerical modeling software [16], as a consequence of the simplicity, there is a possible pay-off to the application scope of the models. In the macro-homogeneous model, the applicability is limited by numerical instabilities when solving the differential equations associated with the modeling, that is, as the current densities grow, the solution profiles abruptly change its gradient in a small region, reaching a behavior similar to singular perturbed differential equations [17], thus this model is precisely used in PEMFC for medium current densities, where the ohmic losses occur. This topic is addressed in the numerical stability discussion of this article, providing directions to improve such stability.

Numerical simulation of a mathematical model implies a set of parameters, but frequently they are not directly obtained, therefore, estimating parameters is necessary to analyze or describe the operation and design of a PEMFC. There are a variety of methods and techniques for parameter estimation, however, algorithms inspired by nature and with stochastic character are widely used due to they are direct search methods that can deal with multiple minima. In addition, they can optimize parameters to improve the energetic performance in a PEMFC. The parameter estimation problem has been tackled by nature inspired algorithms mainly for the semi-empirical model [18,19,20,21], for instance, parameters as maximum current density, active area, equivalent contact resistance to electron conduction and parametric coefficients have been estimated using a genetic algorithm in [2,22,23,24], simulated annealing in [2,25], particle swarm optimization in [26,27], artificial bee swarm in [28], artificial immune system in [29], seeker optimization algorithm in [30], harmonic search in [31], optimization algorithms based on p-systems in [32], differential evolution algorithm in [33].

There exists a diversity of research works related to parameter estimation, so that, modifications have been explored to improve the standard algorithms for PEMFC optimization using different objective functions, for instance, the minimum of the mean of the sum of squared error was used in an adaptation of the Cuckoo Search Algorithm with Onlooker Bee Search (ObCS) in [34]. The squared sum of the errors was used with the Improve Fluid Search Optimization Algorithm (IFSO) to estimate parameters of a PEMFC in [35], this propose simulates Bernoulli's principle in fluid mechanics. Other bio-inspired algorithm is the Chaotic Binary Shark Smell Optimization studied in [36], imitates the ability of the sharks to find their preys incorporate the chaotic movements of the shark in the water, the

objective function given by the minimum of the sum of squared error. Algorithms based on the behaviour of coyotes on the environment is analyzed in [37], this proposal is called Coyote Optimization Algorithm, it uses the objective function as the sum of the absolute error between the experimental data and the simulation of the polarization curve of the PEMFC. Other algorithms based on social behaviour between species as Imperialist competitive algorithm, Shuffled frog-leaping algorithm, and Firefly optimization algorithm are explored in [38], the fitness function is based on minimize the sum of squared errors (SSE) from the output voltage of a PEMFC stack and the estimated voltage given by the semi-empirical model. A recent review that analyze various of the mentioned algorithms is presented in [39].

Fuel cells are part of the renewable energy economy system nested in the energy storage and conversion subsystem [1], therefore, mathematical modeling and simulation can reduce the design costs and assembly times by reducing experimentation in laboratory. In this sense, to optimize the fuel cell performance, it is necessary to have the optimal configuration of the reaction parameters, such as platinum mass, diffusion coefficients, pressures, etc., and design, such as cell dimensions, membrane thickness, etc. [4, 7, 40]. Hence, these parameters can be optimized to obtaining lower manufacturing costs [1] or to improve performance [41, 42]. For instances, in [7], the authors use the macro-homogeneous model to infer that the effective use of platinum catalyst decreases when the current density increases; in [5], the problem of maximizing the current density in a PEMFC at a given electrode potential subject to porosity is approached; in [6] the minimization of activation overpotential for a given current density is presented, and other authors have analyzed the geometry in the flux channels of the gas diffusion layer [9, 10, 11, 1].

In this article, we introduce a unified approach for the numerical simulation and design optimization of a PEMFC. We selected the macro-homogeneous model because of its reliability, precision, and scale of detail. The model is applied for two purposes: a) estimating a set of unknown physical parameters, and b) optimizing a design for improving its performance or minimizing its cost. The objective function is non-linear and non-differentiable, as it is modeled as the sum of squared errors of a discrete polarization curve. Each point in the polarization curve corresponds to the solution of a system of differential equations. To the best of our knowledge there is not other research for optimizing the cell design neither in performance or cost using numerical simulation. For as much as the objective function presents multiple local minima, evolutionary algorithms (EAs) are the appropriate optimizers, considering that they can escape from local minima, do not require derivatives, and are suited for dealing with global optimization problems. Evaluating a candidate solution, that is an individual in the context of EAs, requires of a PEMFC numerical simulation, notice that the simulation software receives random inputs that could lead to numerical failures. In this sense, a contribution of this work is to present a robust simulation algorithm that has been widely stressed by inputting thousands of random configurations that are successfully treated. Another contribution is the comparison of evolutionary algorithms from different families for estimating parameters and optimizing the PEMFC performance. Besides, we introduce objective function definitions for three goals: estimating a set of unknown parameters for reproducing experimental data, improving a base design by maximizing its performance, and reducing the cost via the minimization of the amount

of platinum, without a significant detriment of a base performance. The method is validated by comparing the estimated parameters with the true ones, and by comparing the estimations with reported estimates in the specialized literature. Notice that if the parameters are the same as those reported in the literature and the numerical model reproduces the polarization curve, then the numerical model is valid. Finally, performance optimization and cost minimization are evidenced by comparing the optimized design with a base design.

2 The macro-homogeneous mathematical model

Table 2 Algebraic expressions for the macro-homogeneous mathematical model. Details in [6, 5, 7]

Oxygen equation	$D_{O_2}^{eff} = D_{O_2-m}^{eff} \frac{L_{m,c}}{L_{m,c} + \epsilon_c + L_s} + D_{O_2-w}^{eff} \frac{\epsilon_c}{L_{m,c} + \epsilon_c + L_s}$ $D_{O_2-m}^{eff} = L_{m,c}^{3/2} D_{O_2-m}, \quad D_{O_2-w}^{eff} = \epsilon_c^{3/2} D_{O_2-w}$ $D_{O_2-m} = 1.4276 \times 10^{-11} T - 4.2185 \times 10^{-9}$
Overpotential equation	$\kappa^{eff} = (L_{m,c})^{3/2} \kappa$ $\sigma^{eff} = (1 - L_{m,c} - \epsilon_c)^{3/2} \sigma$
Current density equation	$a = \frac{m_{Pt}}{l_c} A_s, \quad f = \frac{m_{Pt}}{m_{Pt} + m_C}$ $A_s = (227.79f^3 - 158.57f^2 - 201.53f + 159.5) \times 10^3$ $i_0 = 10^{0.03741T - 16.96} \left(\frac{O_2}{C_{O_2,ref}} \right)$

The macro-homogeneous model considers a catalyst layer with a finite thickness in which electrochemical reactions take place, see Figure 2. It is mainly integrated by catalyst clusters of carbon and platinum, a solid GDL phase, a Nafion ionomer phase, and void spaces. Through the CL occurs the diffusion of oxygen, overpotentials, and current [6]. This mathematical model describes the catalyst layer as a one-dimensional problem in a steady state. The membrane and the GDL phases penetrate into the CL, the void spaces are fully flooded by liquid water [6, 8].

The standard mathematical model is described by the following nonlinear boundary value problem [7, 6, 5, 8]:

$$\frac{dO_2}{dz} = \frac{i - I_\delta}{4FD_{O_2}^{eff}} \quad (4)$$

$$\frac{d\eta_{act}}{dz} = \frac{i}{\kappa^{eff}} + \frac{i - I_\delta}{\sigma^{eff}}. \quad (5)$$

$$\frac{di}{dz} = ai_0 \left[\exp\left(\frac{\alpha_c F}{RT} \eta_{act}\right) - \exp\left(\frac{\alpha_a F}{RT} \eta_{act}\right) \right], \quad (6)$$

on $\Omega = [0, l_c]$, with l_c the thickness of the CL, subject to the boundary conditions:

$$\begin{aligned} z = 0, \quad O_2(0) &= O_2^*, \quad i(0) = 0 \\ z = l_c, \quad i(l_c) &= I_\delta, \end{aligned}$$

Fig. 2 The schematic diagram of the catalyst layer.

where the transport due to diffusion of oxygen in the cathode CL following the Fick's law is described in Equation (4), the activation overpotential caused by ohmic losses in Equation (5), and the electrochemical reaction rate following the Butler Volmer equation in Equation (6). In Table 2, we summarize the key algebraic expressions of the system (4) - (6).

2.1 The polarization curve

The polarization curve is a graph associated with the performance of a fuel cell, it presents three characteristic regions (see Figure 3):

- Region 1: Activation losses. Region localized at low current densities which is due to the activation losses or kinetic activation of the electrochemical reaction that occurs at the catalytic layers. Its shape is due to the exponential voltage drop for low current densities.
- Region 2: Ohmic losses. It is observed in the zone of moderate current densities and it has a linear trend due to electronic resistances or ionic mass transport losses.
- Region 3: Concentration losses. Region observed at high current densities due to the resistance to the mass transfer of oxygen from the bulk to the active sites localized in the catalytic layer. Its shape is characterized by the rapid drop in voltage until it reaches the value of 0 V.

The polarization curve for the macro-homogeneous mathematical model is described by (7) according to [6], in this paper it is used for characterizing the cell, the parameters are estimated to approximate an experimental polarization curve, and they are optimized to increase the polarization curve area or to maintain a similar polarization curve while minimizing the platinum mass loading.

$$V_{FC} = E_{Nerts} - \eta|_{z=L_c} - R_{Ohmic}I_\delta. \quad (7)$$

where R_{Ohmic} is the specific ohmic resistance of the cell and E_{rev} is the reversible cell voltage [5,6],

Fig. 3 Polarization curve for a PEMFC.

$$\begin{aligned}
 E_{Nerts} &= E^0 - \frac{RT}{nF} \ln [P_{H_2} P_{O_2}^{0.5}] \\
 &= 1.229 - 0.85 \times 10^{-3} (T - 298.15) \\
 &\quad + 4.31 \times 10^{-5} T [\ln (P_{H_2}) + 1/2 \ln (P_{O_2})]
 \end{aligned} \tag{8}$$

3 Reformulation of the macro-homogeneous model as singularly perturbed ODE systems

On one hand, a drawback of the macro-homogeneous model is the numerical instabilities, mainly in the *concentration losses* region (Figure 3). On the other hand, a requirement for applying bio-inspired algorithms is to have a reliable numerical simulator, considering that the simulator is stressed by inputting thousands of random parameters, and it must deliver precise numerical solutions for each input. Hence, in this section, we revisit the non-linear coupled system of the PEMFC to improve its numerical stability.

The most common coupled system of nonlinear ordinary differential equations consists of a problem with the three equations of the system (4), (5), (6), the system of ODEs formulated as initial value problem, is usually solved using standard numerical techniques as Runge-Kutta methods [5,7,6,8]; with these methods the system presents numerical stability if I_δ belongs to Region 1 and Region 2 of the polarization curve, however, the solvers become unstable if I_δ belongs to Region 3 (Concentration losses, see Figure 3).

To obtain more realistic solutions, the macro-homogeneous system should be formulated as a boundary value problem [6,8], where we need to estimate the initial value condition for Equation (5) (overpotential).

As it is described in [5,6,7] Equation (4), is obtained from,

$$-D_{O_2}^{eff} \frac{d^2 O_2}{dz^2} = -\frac{1}{4F} \frac{di}{dz}, \tag{9}$$

with the boundary values,

$$z = 0, \quad O_2(0) = O_2^*, \quad z = l_c, \quad \frac{dO_2(l_c)}{dz} = 0, \quad (10)$$

From physical experiments in PEMFC it has been established that $0 < D_{O_2}^{eff} \ll 1$, [43,44], then Equation (9) is a second order singularly perturbed differential equation, and its numerical implementation is important to obtain regularized solutions. To address this issue, we introduce two approaches for improving the numerical stability of the system, the first is rewriting Equation (4) using the change of variable given by subsystem (11),

$$\frac{dO_2}{dz} = -\frac{N_{O_2}}{D_{O_2}^{eff}}, \quad \frac{dN_{O_2}}{dz} = -\frac{1}{4F} \frac{di}{dz}, \quad (11)$$

Then, the second approach is given by using $D_{O_2}^{eff}$ in the second equation, rewriting Equation (11) as Equation (12),

$$\frac{dO_2}{dz} = -N_{O_2}, \quad \frac{dN_{O_2}}{dz} = -\frac{1}{4FD_{O_2}^{eff}} \frac{di}{dz}. \quad (12)$$

On each subsystem, we have a first order singularly perturbed ODE system. First, subsystem (11) is more numerically stable than the standard Equation (4). To clarify this, we identify the slow variable as N_{O_2} , and the fast variable as O_2 , now, we consider the change of variable $z = D_{O_2}^{eff} \tau$, with $\tau \in [0, l_c/D_{O_2}^{eff}]$, then we have a new formulation,

$$\frac{dO_2}{d\tau} = -N_{O_2}, \quad \frac{dN_{O_2}}{d\tau} = -\frac{1}{4F} \frac{di}{d\tau}, \quad (13)$$

With this change of variable, called homogenization [45,46], the diffusive coefficient disappears, however, equations become completely determined by the slopes N_{O_2} and $\frac{di}{d\tau}$, it is important to note that these profiles have fast growth in a small subdomain Ω_f of Ω , this implies that there exists a dependency of the solutions with the correct approximations of N_{O_2} and $\frac{di}{d\tau}$.

With the second proposed subsystem (12), we use a similar analysis and we use the same change of variable transforming it into,

$$\frac{dO_2}{d\tau} = -D_{O_2}^{eff} N_{O_2}, \quad \frac{dN_{O_2}}{d\tau} = -\frac{1}{4FD_{O_2}^{eff}} \frac{di}{d\tau}. \quad (14)$$

In this case, we observe that the coefficient $D_{O_2}^{eff}$ appears in the two equations, this compensates the effect of the fast growth of the slope N_{O_2} providing more numerical stability for the state variable O_2 . With this last formulation, we obtain the highest numerical stability among the three models. Despite the reformulation of the system of equations, a robust and stable numerical method is necessary, due to the complexity of the combinations of values in the parameters that are evaluated when applying the optimization methods. The system of ODEs in this work are solved using the Runge-Kutta with an implicit Lobatto technique [47].

Fig. 4 Comparative results of $i(z)$ using: a) system Mod_1 - green solid line, b) system Mod_2 - red dotted line, c) system Mod_3 - blue solid dotted line. The actual expected solution is similar to those of Mod_1 and Mod_2

Fig. 5 Close up to Figure 4 for the comparison of the solutions of $i(z)$ using: a) system Mod_1 - green solid line, b) system Mod_2 - red dotted line, c) system Mod_3 - blue solid dotted line. At this scale it is clear that only Mod_3 provides more numerical stability.

To demonstrate the numerical stability of our proposals, we carried out numerical experiments with the three systems according to the following,

- Mod_1 , is the formulation of the system solved in the following order, Equation (4), Equation (5), and Equation (6). This is the most widely used system in the literature [6,5,7].

- Mod_2 , is the first formulation introduced in this article, solved in the following order, System (11), Equation (5), and Equation (6).
- Mod_3 , is the second formulation introduced in this article, solved in the following order, System (12), Equation (5), and Equation (6).

The boundary conditions for Mod_2 and Mod_3 are:

$$\begin{aligned} z = 0, \quad O_2(0) = O_2^*, \quad i(0) = 0 \\ z = l_c, \quad N(l_c) = 0, \quad i(l_c) = I_\delta, \end{aligned}$$

In Figure 4, we present the results of the current density $i(z)$ using the boundary condition $i(l_c) = 9672 \text{ mAcm}^{-2}$ for the three systems. The expected solution curve for the current density must increase within a small interval close to zero tending to 9672 and must stabilize at that value, thus, the solution obtained with the system Mod_1 presents an erroneous profile.

Figure 4 shows that the red dotted line (Mod_2) presents the expected curve profile, hence the result from system Mod_2 is more numerically stable than that from system Mod_1 . Nevertheless, in Figure 5, in a close up of the solutions, we notice that Mod_1 , the solid green line, presents the largest abrupt changes, while Mod_2 presents small variations around 9672 mAcm^{-2} , finally, the solution using system Mod_3 (blue solid-dotted line) avoid any numerical issue in contrast with those obtained from systems Mod_1 and Mod_2 . Then, the solution of $i(z)$ using system (12), labeled as Mod_3 is the most stable of the three approaches, and closely reproduces the expected polarization curve.

4 Objective functions and algorithms for parameter estimation

Nature-inspired algorithms are a type of meta-heuristics [48] that emulate the behavior of species in nature to improve a fitness function. Regarding the family of the algorithm, they emulate different natural processes and, as a consequence, the performance varies from problem to problem from one family to another. This paper is devoted to three of the most successful ones: the Genetic Algorithm (GA) [49, 22] emulates the evolution of a population of individuals that compete for inheriting their genes, the fittest are selected for reproduction via recombination of genes, in this case, such genes are the parameters of the PEMFC and the fitness is the error to the experimental data. The Particle Swarm Optimization (PSO) [26] emulates swarms behavior such as flocks of birds, the particles are updated using a velocity which is a combination of information of the best particle in a neighborhood that is defined as a graphical model, the best global particle and inertial vectors. The swarm of particles move through the landscape fitness, then, they concentrate the search in the most promising region. Finally, Estimation of Distribution Algorithms (EDAs) [50] were derived from statistical and probabilistic modeling of GAs, they estimate a search probability distribution and sample new candidate solutions. Even though EDAs address the same kind of problems as other nature-inspired algorithms, they present additional benefits, such as convergence to the best solution known, and the possibility of incorporating problem specific knowledge or search bias, this promotes the search around the local and possible global minimum, and circumvents to set arbitrary stopping criteria to the algorithm.

All the algorithms require as main input an objective function to minimize/maximize. In this section, we introduce three objective functions and a brief recall of the optimization algorithms used for approximating the optimum. The first objective function is used for estimating unknown parameters, the second is the performance maximization and the third is the minimization of the platinum mass loading m_{Pt} , the last two objective functions are part of the contributions of this article.

4.1 Objective functions

4.1.1 Parameter estimation problem

Given a set of unknown parameters $\vec{\theta}$, we define the objective function, $SSE : \Theta \rightarrow \mathbb{R}_+$, as the sum of the squared error between experimental data obtained from a polarization curve and the output of the model,

$$\min SSE(\theta) = \sum_{i=1}^m \left| V_i^{FC}(z_i) - V(z_i; \vec{\theta}) \right|^2, \quad (15)$$

where m is the number of the experimental data, V_i^{FC} and $V(z_i; \vec{\theta})$ are the i -th experimental datum and the voltage output of the macro-homogeneous model, respectively, at current density z_i . The parameter estimation problem consists of determining the set $\vec{\theta} \in \Theta$ that returns a minimum value for $SSE(\vec{\theta})$.

4.1.2 Performance maximization problem

We consider to maximize the voltage generated by the cell according to the polarization curve, then this problem is equivalent to minimize the difference of an ideal voltage to the actual voltage delivered by the numerical simulation, see Figure 3, thus, the second optimization problem is in Equation (16).

$$\min SSE_2(\theta) = \sum_{i=1}^m \left| 1.2 - V(z_i; \vec{\theta}) \right|^2. \quad (16)$$

Where z_i is a current density value and $\vec{\theta}$ are the parameters of the fuel cell. Notice that the ideal voltage, 1.2 V in this case, means that the cell maintains a maximum constant voltage in all the regions of the polarization curve, that is impossible, but the optimization problem looks for the cell that best approximates this ideal performance.

4.1.3 Minimization of the platinum mass loading

An objective function to minimize the platinum mass loading, and consequently, the cost of the cell, follows the idea of generating a polarization curve with the same area than the experimental polarization curve, this is defined with the first term of Equation (17), where $d_A = (A_{sim} - A_{exp})/A_{exp}$, A_{sim} is the simulated polarization curve, and A_{exp} is the experimental polarization curve. The second term is the variable $m_{Pt} \in [L_{m_{Pt}}, U_{m_{Pt}}]$ that is the platinum mass loading,

$$\min F_{obj} = \frac{m_{Pt}}{U_{m_{Pt}} - L_{m_{Pt}}} + e^{|d_A|}. \quad (17)$$

With this formulation, we preserve the performance while minimize the platinum mass loading.

4.2 Genetic algorithm

Genetic algorithms are based on the processes of biological evolution, they evolve a population of individuals that are a codification of candidate solutions. The population evolves via crossover, mutation, and selection operators, the crossover preserves genetic information, that is to say, it recombines existing partial solutions, the mutation applies small perturbations to instances, generating values that do not depend on the current population, and the selection defines the fittest individuals for reproduction, biasing the search to the most promising regions. In each iteration, the algorithm selects a set of the best fitted individuals according to their fitness function, these are the parents, who generate individuals with expected better qualities called children, through the generations the population evolves until a stopping criterion is reached. We use the GA according to [51], this version is as follows:

- Generate a random population and compute their fitness based on the objective function.
- Score the elements of the population follow the fitness values.
- Scale the fitness to obtain the expectation value.
- Select the parents based of a selection criterion and the expectation value.
- Select a elite subset of the current population based on the best expectation values, and pass them to the next generation.
- Create a children population from the selected parents population using the mutation and crossover operators.
- Replace the old population with elite and children populations.
- The algorithm stops until the stopping criteria is reaching.

4.3 Particle Swarm Optimization

Particle swarm optimization is an algorithm inspired by the social behavior of animals, such as ants, fish, birds, etc. The algorithm is based on the criterion of the existence of an "invisible" method to share information between unstructured and generally dispersed biological groups. It emulates the interaction between the members of a biological group [52].

The PSO version used here [27] is based on the algorithm described in [53], using modifications suggested in [54] and in [55]. In general, the algorithm works as follows:

- Create an initial swarm of particles, and assign initial velocities.
- Evaluates the objective function at each particle location.
- Determines the best location and the best (lowest) function value.

- Computes new velocities, using the current velocities, the particle best location, and the best location of their neighbors.
- Until the algorithm reaches a stopping criterion, updates the particle positions (the new position is the old one plus the randomly scaled velocity), velocity, and neighbors.

4.4 The Univariate Marginal Distribution Algorithm with Gaussian Models

The Estimation of Distribution Algorithms (EDAs) were derived from the probabilistic modeling of Genetic Algorithms [56,57], are based on the dynamic of populations where the offsprings of the generations are generated by sampling from a probabilistic distribution model, described by the parents set. Hence, the learning of these algorithms comes from estimating a parametric probability distribution from the selected set, intending to associate the highest probability to the most promising regions. This is different from other nature-inspired algorithms in which the evolution of the populations is carried out by crossover and mutation operators [56,57,58,59,60,61,62].

The Univariate Marginal Distribution Algorithm with Gaussian models (*UMDA^G*) is a kind of EDA [63,64,65,66,67,21] that assumes that the probabilistic distribution of the populations is a l -dimensional joint probability function that is factored as a product of l univariate and independent probability functions. Then, at each iteration, the algorithm compute the join probability given by

$$\mathbf{P}_k(\vec{\theta}_{k,h}|S_{\theta_k}) = \mathbf{P}_k(\theta_{k,h}^1, \theta_{k,h}^2, \dots, \theta_{k,h}^l|S_{\theta_k}) = \prod_{i=1}^l \mathcal{P}_{k,i}(\theta_{k,h}^i|S_{\theta_k}). \quad (18)$$

Each function $\mathcal{P}_{k,i}(\theta_{k,h}^i)$ is a normal distribution function with mean and variance,

$$\mu_{k,i}(S_{\theta_k}) = \sum_{s=1}^{N_{ind}} \theta_{k,s}^i p_{k,i}(\theta_{k,s}^i), \quad \sigma_{k,i}^2(S_{\theta_k}) = \sum_{s=1}^{N_{ind}} (\mu_{k,i} - \theta_{k,s}^i)^2 p_{k,i}(\theta_{k,s}^i), \quad (19)$$

where $p_{k,i}(\theta_{k,s}^i|S_{\theta_k})$ are weights associated with the N_{ind} individuals, for $0 < i \leq l$.

In the algorithm, at each iteration, the weights $p_{k,i}(\theta_{k,s}^i|S_{\theta_k})$ are computed using a threshold ν and a selected subset $S_{\theta_k} \subset \theta_k$, this is called the truncation method [50]. The assigned probability to each $\theta_{k,s}^i$ in the iteration k , for $0 < i \leq l$ and $\vec{\theta}_{k,s} \in \theta_k$ is computed following the function of the threshold ν ,

$$p_{k,i}(\theta_{k,s}^i|S_{\theta_k}) = \begin{cases} \frac{1}{\nu}, & \text{if } \vec{\theta}_{k,s} \in S_{\theta_k}, \\ 0, & \text{if } \vec{\theta}_{k,s} \notin S_{\theta_k}. \end{cases} \quad (20)$$

The stop criteria is a function of the norm of the normalized variance of the optimized parameters [21],

$$norm_k = \left\| \left\{ \frac{var(\theta_i)}{U_Bound(\theta_i) - L_Bound(\theta_i)} \right\}_{i=1}^l \right\|. \quad (21)$$

This criterion provides a natural way to stop the algorithm compared with the standard criteria as a function of the number of generations.

5 Results

	Data set 1				Data set 2			Data set 3		
	DO_{2gdl}	m_{pt}	p_{pt}	ϵ_c	m_{pt}	p_{pt}	ϵ_c	m_{pt}	m_C	ϵ_c
Lim_{inf}	0.2	0.2	21	0.31	0.3	18	0.3	0.5	0.91	0.3
Lim_{sup}	0.4	0.4	22	0.35	0.4	22	0.5	0.7	0.93	0.5

Table 3 Upper and lower bounds for parameter estimation.

Parameters	Reported	UMDA ^G	GA	PSO
Dataset 1				
$DO_{2gdl}(cm^2/s)$	0.2396 ^a	0.24127 <i>pm</i> 1.223e-03	0.23765 <i>pm</i> 1.411e-16	0.23923 <i>pm</i> 1.693e-16
$m_{Pt} (mg/cm^2)$	0.332 ^a	0.33178 <i>pm</i> 1.215e-03	0.33006 <i>pm</i> 2.200e-18	0.33159 <i>pm</i> 1.700e-19
$\rho_{Pt} (g/cm^3)$	21.5 ^a	21.928 <i>pm</i> 0.11694	21.999 <i>pm</i> 3.613e-15	21.757 <i>pm</i> 7.226e-15
ϵ_c	0.33 ^a	0.32667 <i>pm</i> 2.68e-03	0.33722 <i>pm</i> 5.646e-17	0.33152 <i>pm</i> 1.000e-20
SSE	–	7.9047e-08 <i>pm</i> 1.442e-07	2.5863e-08 <i>pm</i> 1.100e-20	1.0377e-09 <i>pm</i> 1.000e-20
Dataset 2				
$m_{Pt} (mg/cm^2)$	0.35 ^b	0.35009 <i>pm</i> 1.0709e-3	0.35472 <i>pm</i> 2.82e-16	0.34129 <i>pm</i> 1.129e-16
$\rho_{Pt} (g/cm^3)$	21.4 ^b	20.0356 <i>pm</i> 0.35619	18.6551 <i>pm</i> 3.61e-15	20.0871 <i>pm</i> 1.445e-14
ϵ_c	0.4 ^b	0.39818 <i>pm</i> 1.4348e-2	0.32348 <i>pm</i> 2.82e-16	0.5000 <i>pm</i> 1.000e-20
SSE	–	2.348e-17 <i>pm</i> 3.969e-17	3.900e-16 <i>pm</i> 1.0e-20	1.989e-15 <i>pm</i> 1.000e-20
Dataset 3				
$m_{Pt} (mg/cm^2)$	–	0.50053 <i>pm</i> 1.0226e-03	0.50006 <i>pm</i> 2.258e-16	0.50000 <i>pm</i> 1.000e-20
$m_C (g/cm^3)$	–	0.91003 <i>pm</i> 8.7887e-05	0.910006 <i>pm</i> 3.389e-16	0.91000 <i>pm</i> 1.100e-20
ϵ_c	–	0.41802 <i>pm</i> 4.4027e-04	0.417761 <i>pm</i> 1.000e-20	0.41782 <i>pm</i> 1.693e-16
SSE	–	0.21417e-02 <i>pm</i> 4.741e-06	0.21290e-02 <i>pm</i> 1.32e-18	0.21287e-02 <i>pm</i> 8.80e-19

Table 4 Comparative results for parameter estimation with reference values from [5]^a and [6]^b.

In this section, we use the evolutionary algorithms presented in Section 4 applied to the macro-homogeneous model in different scenarios. In subsection 5.1, we used algorithms to estimate a set of parameters to reproduce the experimental

polarization curve. In subsection 5.2, we present the optimization to increase the voltage generated by the cell and the minimization of the platinum mass loading used in the cathode.

5.1 Validation for parameter estimation

This section presents comparative optimizations with three experimental datasets: Song *et al.* (2004) [5], Dalasm *et al.* (2010) [6] and Salva *et al.* (2016) [68]. Using these datasets, we estimate parameters for reproducing the polarization curve with the three optimization algorithms: GA, PSO, and $UMDA^G$. We carried out 30 executions of each algorithm in order to obtain statistical results for comparing their performance. For all simulations, we set the parameters for GA and PSO as suggested by MATLAB [51, 27]. For the three algorithms (GA, PSO, $UMDA^G$), we fixed population size with 100 individuals (particles), the number of generations in $200nvars$, where $nvars$ is the number of variables, and tolerance is 10^{-6} .

The choice of parameters to estimate in this work follows previous studies in [6, 5], where the authors analyze the feasibility of varying some parameters of the macro-homogeneous model, such as m_{pt} and ϵ_c , in the performance of the polarization curve. The upper and lower bounds are derived of an analysis of the results obtained in [6, 5, 7]. For estimating the initial boundary condition $\eta(0) = \eta_1$ for Equation (5) the shooting method is applied using the MATLAB function `bvp5c`, see [47].

5.1.1 Dataset 1: Song *et al.* (2004)

The first dataset is obtained from the research of Song. *et al.* (2010) [5], the experimental polarization curve for this dataset presents a whole desirable polarization curve as in Figure 3. We estimate the set of parameters given by $\theta = \{\epsilon_c, m_{Pt}, \rho_{Pt}, D_{O_2, GDL}^{eff}\}$, the first three parameters was selected due we consider that are important in the modeling, $D_{O_2, GDL}^{eff}$ appears in the initial condition for state variable O_2 , describes the effective diffusion coefficient of oxygen in GDL, more details in [5]. The values of the base parameters of the macro-homogeneous model are presented in 7.

The upper and lower bounds for searching each parameter are in Table 3, these intervals are set according to the results reported in [5]. In Table 4, the statistical results obtained from the estimation of parameters are presented by applying the three evolutionary algorithms, it can be seen that the results are similar with order of approximation for the SSE of less of 10^{-7} . In Figure 6, we plotted the polarization curve of the best solution for each estimate. In Figure 6-b), we observe that the best approximation to the experimental data is that delivered by the $UMDA^G$, but the standard deviation of its 30 executions ($\approx 10^{-7}$) is greater than the standard deviation obtained with the PSO that is 10^{-20} .

5.1.2 Dataset 2: Dalasm *et al.* (2010)

The second dataset is obtained from [6], describes regions 1 and 2 of the ideal polarization curve (see Figure 3), originally, the experimental data are from Ticianelli

Fig. 6 a) Comparative plots of the polarization curves with the best set of estimated parameters for the UMDA^G, GA, and PSO for data from [5]; b) Zoom of the approximations at $i = 0.35 Acm^{-2}$.

et al. [69]. We select to estimate the set of parameters given by $\theta = \{m_{Pt}, \rho_{Pt}, \epsilon_c\}$, the choice of these parameters, upper and lower bounds for each parameter (Table 3), were considered following previous results in [6]. The values of the complete base parameters used in [6] are presented in 8.

Comparative statistical results are presented in Table 4, the results show that the estimations have a SEE around the order of less of 10^{-17} for each algorithm. In Figure 7, we plotted the polarization curve of the best solution for each estimate. In this case, the $UMDA^G$ has a mean in the three parameters that is closer to the reported values, compared to the means of each parameter obtained by GA and PSO.

5.1.3 Data set 3: Salva et al. (2016)

The third dataset is taken from [68], in this case, the authors only report the experimental data without performing mathematical modeling, therefore, in this work we try to adjust the macro-homogeneous model to the experimental data, thus providing a model with the adjusted parameters that can reproduce it. We estimate the loads of platinum and carbon, and the porosity of the CL, then $\theta = \{m_{Pt}, m_c, \epsilon_c\}$. The proposed values of the base parameters used in this work for the macro-homogeneous model are shown in 9. Upper and lower bounds for the search space for optimization are in Table 3.

Comparative plot of the best parameter estimations are shown in Figure 8, we can observe that the polarization curves are similar for the three algorithms. Table 4, show statistical results, observing that the parameters reaching a stable values of SSE around 0.21417 for the three algorithms with standard deviation of less of 10^{-6} .

5.2 Performance maximization and platinum mass loading minimization

In the previous section, we have shown that the three comparing algorithms perform adequately for the three datasets, nevertheless the experimental dataset that is reproduced with the highest accuracy is the dataset 2 of Dalasm [6]. Hence, in this section we optimize the cell performance according Equation (16) and the platinum mass loading according to Equation (17), in both cases we use the dataset of Dalasm [6] regarding that the model closely reproduces the experimental data, and we apply the three comparing algorithms considering that all of them are statistically consistent and deliver a low error.

5.2.1 Performance maximization

We aim to optimize the performance requiring to the PEMFC to deliver a voltage as close as possible to the ideal voltage of $E = 1.2$, that is to say, the polarization curve initially have a voltage around 1, it decreases when the current density increases, hence we introduce an ideal voltage of 1.2 V that is greater than any actual voltage delivered by the cell. The objective function measures the distance between the actual model output and the ideal voltage, according to Equation (16), the lower the distance is, the greater the voltage delivered. The upper and lower bounds are presented in Table 3 (Dataset 2).

The parameter values after optimization for the three algorithms are shown in Table 5. Figure 9 compares a base design with the optimized performance of the three algorithms, Sim_{ref} , was obtained with the best parameter optimization of the $UMDA^G$, $[m_{pt}, p_{pt}, \epsilon_c] = [0.349997, 19.603549, 0.399814]$. Results of the algorithms

Fig. 7 a) Comparative plots of the polarization curves with the best set of estimated parameters for the UMDA^G, GA, and PSO for data from [6]; b) Zoom of the approximations at $i = 2000 m A cm^{-2}$

shown that the optimized values consistently are around to $m_{pt} = 0.4$, $p_{pt} = 22$, $\epsilon_c = 0.5$.

For the sake of completeness, the voltage for each current density value is increased at least 0.42% and up to 0.44%. The total voltage gain is 4.26% with respect to the base polarization curve.

Fig. 8 a) Comparative plots of the polarization curves with the best set of estimated parameters for the UMDA^G, GA, and PSO for data from [68]; b) Zoom of the approximations at $i = 3600 mAc m^{-2}$

The results validate that the same methodology and numerical modeling with a different objective function are successfully applied for both purposes: estimating unknown parameters and optimizing the design performance.

Fig. 9 a) Comparative plots of the performance optimization with Equation (16). The closer to E_0 (horizontal line) a curve is, the best performance the cell has. The red solid line is the base design. The blue dashed-dashed line is the $UMDA^G$, green dashed-dotted line is the GA, and the PSO optimization is the magenta dotted-dotted line. b) Zoom at $i = 4000 mA cm^{-2}$. The three optimized cells deliver a higher voltage than the base design.

5.2.2 Minimization of platinum loading m_{Pt}

To minimize the platinum loading, we use the objective function given by Equation (17). The search limits are set regarding the ranges of values reported in the specialized literature, with the purpose of searching on the largest feasible range. The lower bound of the search limits for m_{Pt} is set to 0.2 as it is suggested in

Fig. 10 a) Comparative plots of the polarization curves optimizing m_{Pt} with the three algorithms: base design - black solid line, optimization with objective function (15), $UMDA^G$ - blue dashed-dashed line, GA - green dashed-dotted line, and PSO - red dotted-dotted line; b) Zoom of the approximations at $i = 300 mAc m^{-2}$

[70], the upper bound is set to 0.5, that is the average of the values suggested in [6]. The lower bound for ϵ_c is 0.2, that is the suggested value in [5], and the upper bound is 0.5 that is the suggested value in [6]. Finally, the search range for p_{pt} are [15,24] to have sufficient flexibility for compensating for the possible reduction of m_{Pt} . The optimum approximations are presented in Table 6. The GA

	m_{pt}	p_{pt}	ϵ_c	SSE_2
$UMDA^G$	0.3999985	21.9999997	0.4999997	2.184322
GA	0.3994409	21.23680	0.500000	2.184321
PSO	0.400000	22.00000	0.500000	2.184321

Table 5 Parameter values from the performance optimization, SSE_2 is the objective value according to Equation (15).

and PSO provides very similar sets of values. The best results reduce the quantity of platinum by 42% while the voltage difference is in average less than 1.5% in contrast to the base design. That is to say, the optimized cell delivers almost the same voltage than the base design while the most expensive component has been significantly reduced. Figure 10 compares the polarization curves respect to the base design, the polarization curves are very similar for the three algorithms. Then, the reported values for a low-cost cell design are approximately $m_{pt} = 0.2$, $p_{pt} = 24$, and $\epsilon_c = 0.5$. An important result is that the optimized value of m_{pt} belongs to the range of $0.20 \pm 0.05 \text{ mg cm}^2$, that is congruent with the reported experimentally in [70].

	m_{pt}	p_{pt}	ϵ_c	F_{obj}
$UMDA^G$	0.200297051	20.700952726	0.4999999995	2.2342745119234
GA	0.200000000	23.999999466	0.5000000000	2.2333318027298
PSO	0.200000000	24.000000000	0.5000000000	2.2333318027287

Table 6 Optimum approximations for the platinum mass loading optimization according to Equation (16).

6 Conclusion

In this paper, we introduce a method that combines stable numerical simulation and optimization for addressing three problems. The first is parameter estimation of unknown parameters to reproduce experimental data, the second is the optimization of the performance by maximizing the voltage output, and the third is the minimization of the platinum mass loading while maintaining similar performance to the base design.

As a first contribution, we introduce a rearrangement of the governing system of ordinary differential equations that significantly reduces the numerical instabilities, this issue is essential for applying nature-inspired algorithms, considering that we must evaluate a huge amount of random inputs. A second contribution is the adequate modeling of the objective functions. Notice that assembling the numerical simulator with the evolutionary algorithms is straightforward, nevertheless, the quality of the results depends on the objective function model. Our results show that the bio-inspired algorithms solved the parameter estimation problem modeled as a sum of squared errors, in Equation (15), in consideration of these results, we

model the performance optimization, in Equation (16), as a particular case of the parameter estimation problem but using a constant target polarization curve, the numerical experiments demonstrate that this model is adequate because we improve the base design of the cell 4.26%. To the best of our knowledge, there are no results reporting the minimization of the platinum mass loading with the stable macro-homogeneous model. Hence, we introduce this problem considering that one of the most important limitations for the popularization of fuel cells is their cost in contrast with other green technologies. In this regard, we propose an objective function that measures the area between the simulated polarization curve and the base design, in other words, we reduce the cost maintaining a similar performance, achieving a platinum mass loading reduction of 42%.

Regarding the optimization algorithms, all of them return similar approximations, but the PSO consistently delivers the best objective values. Nevertheless, the nature of the probabilistic construction of the *UMDA*^G produces a region of confidence, that is to say, for this algorithm we know that the results are the best from a normal probability distribution with a small variance.

Finally, we conclude that a first step for applying stochastic optimization methods to the parameter estimation and design optimization of fuel cells is to develop a numerically stable simulation method. Another conclusion is that a similar assembly of optimizer and simulator can be applied to a variety of problems, for instance, for parameter estimation or performance optimization, if the optimization problem is modeled adequately.

Declarations

Competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Availability of data and material

The input data and results of the executions are in https://github.com/pemfc-team-CMCGCY/MH_results.

Author contributions

All authors contributed to the study Conceptualization. Supervision, Funding acquisition, and Writing-review & editing were performed by Salvador Botello. Supervision, Methodology and Writing-review & editing were performed by S. Ivvan

Valdez. Investigation, Validation, and Writing-review & editing were performed by L. C. Ordoñez. Investigation, Software, Visualization and Writing-original draft were performed by Luis Blanco-Cocom.

References

1. J. Wang, H. Wang, Y. Fan, *Engineering* **4**(3), 352 (2018). DOI <https://doi.org/10.1016/j.eng.2018.05.007>. URL <https://www.sciencedirect.com/science/article/pii/S2095809917307750>
2. M. Outeiro, R. Chibante, A. Carvalho, A. de Almeida, *Journal of Power Sources* **185** (2), 952 (2008)
3. S. Sun, Y. Su, C. Yin, K. Jermsittiparsert, *Energy Reports* **6**, 1501 (2020). DOI <https://doi.org/10.1016/j.egy.2020.06.002>. URL <http://www.sciencedirect.com/science/article/pii/S2352484720306697>
4. M. Secanell, K. Karan, A. Suleman, N. Djilali, *Electrochim. Acta* **52**, 6318 (2007)
5. D. Song, Q. Wang, Z. Liu, T. Navessin, M. Eikerling, S. Holdcroft, *Journal of Power Sources* **126**(1), 104 (2004). DOI 10.1016/j.jpowsour.2003.08.043
6. N. Khajeh-Hosseini-Dalasm, M. Kermani, D.G. Moghaddam, J. Stockie, *International Journal of Hydrogen Energy* **35**, 2417 (2010)
7. C. Marr, X. Li, *Journal of Power Sources* **77**(1), 17 (1999). DOI [https://doi.org/10.1016/S0378-7753\(98\)00161-X](https://doi.org/10.1016/S0378-7753(98)00161-X). URL <http://www.sciencedirect.com/science/article/pii/S037877539800161X>
8. H. Heidary, M. Jafar Kermani, N. Khajeh-Hosseini-Dalasm, *International Journal of Hydrogen Energy* **41**(47), 22274 (2016)
9. D. Lu, N. Djilali, T. Berning, *Journal of Power Sources* **106**(1-2), 284 (2002). ISI:000175342800040
10. T. Berning, N. Djilali, *Journal of The Electrochemical Society* **150**(12), A1589 (2003). DOI 10.1149/1.1621412. URL <https://doi.org/10.1149/1.1621412>
11. T. Berning, N. Djilali, *Journal of Power Sources* **124**(2), 440 (2003). DOI [https://doi.org/10.1016/S0378-7753\(03\)00816-4](https://doi.org/10.1016/S0378-7753(03)00816-4). URL <http://www.sciencedirect.com/science/article/pii/S0378775303008164>
12. W. Tiedemann, J. Newman, *Journal of The Electrochemical Society* **122**(11), 1482 (1975). DOI 10.1149/1.2134046
13. L. You, H. Liu, *International Journal of Hydrogen Energy* **26**(9), 991 (2001). DOI [https://doi.org/10.1016/S0360-3199\(01\)00035-0](https://doi.org/10.1016/S0360-3199(01)00035-0). URL <http://www.sciencedirect.com/science/article/pii/S0360319901000350>
14. A. Shah, G.S. Kim, P. Sui, D. Harvey, *Journal of Power Sources* **163**(2), 793 (2007). DOI <https://doi.org/10.1016/j.jpowsour.2006.09.022>. URL <http://www.sciencedirect.com/science/article/pii/S0378775306019264>. Selected Papers presented at the FUEL PROCESSING FOR HYDROGEN PRODUCTION SYMPOSIUM at the 230th American Chemical Society National Meeting Washington, DC, USA, 28 August – 1 September 2005
15. G. Wang, P.P. Mukherjee, C.Y. Wang, *Electrochimica Acta* **52**(22), 6367 (2007). DOI <https://doi.org/10.1016/j.electacta.2007.04.073>. URL <http://www.sciencedirect.com/science/article/pii/S0013468607005683>
16. M. Secanell, A. Jarauta, A. Kosakian, M. Sabharwal, J. Zhou, *PEM Fuel Cells, Modeling* (Springer New York, New York, NY, 2017), pp. 1–61. DOI 10.1007/978-1-4939-2493-6_1019-1
17. N. Kopteva, E. O'Riordan, *International Journal of Numerical Analysis and Modeling* **7**(3), 393 (2010). DOI <https://doi.org/>
18. K. Priya, K. Sathishkumar, N. Rajasekar, *Renewable and Sustainable Energy Reviews* **93**, 121 (2018). DOI <https://doi.org/10.1016/j.rser.2018.05.017>. URL <http://www.sciencedirect.com/science/article/pii/S1364032118303642>
19. Y. Chen, N. Wang, *International Journal of Hydrogen Energy* **44**(5), 3075 (2019). DOI <https://doi.org/10.1016/j.ijhydene.2018.11.140>. URL <http://www.sciencedirect.com/science/article/pii/S0360319918337455>
20. A. Askarzadeh, L. dos Santos Coelho, *International Journal of Hydrogen Energy* **39**(21), 11165 (2014). DOI <https://doi.org/10.1016/j.ijhydene.2014.05.052>. URL <http://www.sciencedirect.com/science/article/pii/S0360319914014001>

21. L. Blanco-Cocom, S. Botello-Rionda, L. Ordoñez, S.I. Valdez, *Mathematics and Computers in Simulation* **185**, 218 (2021). DOI <https://doi.org/10.1016/j.matcom.2020.12.021>. URL <http://www.sciencedirect.com/science/article/pii/S0378475420304766>
22. M. Ohenoja, K. Leiviskä, *International Journal of Hydrogen Energy* **35**(22), 12618 (2010). DOI <https://doi.org/10.1016/j.ijhydene.2010.07.129>. URL <http://www.sciencedirect.com/science/article/pii/S036031991001517X>. Bio-Ethanol and Other Renewable Sources and Reforming Process for Sustainable Hydrogen Production
23. Z.J. Moo, X.J. Zhu, L.Y. Wei, G.Y. Cao, *Int J Energy Res* **20**, 585 (2006)
24. L. Zhang, N. Wang, *International Journal of Hydrogen Energy* **38** (1), 219 (2013)
25. M.T. Outeiro, R. Chibante, A.S. Carvalho, A.T. de Almeida, *International Journal of Energy Research* (2009)
26. M. Ye, X. Wang, Y. Xu, *International Journal of Hydrogen Energy* **34**(2), 981 (2009). DOI <https://doi.org/10.1016/j.ijhydene.2008.11.026>
27. S. Mirjalili, *MATLAB Central File Exchange* (2020). URL <https://www.mathworks.com/matlabcentral/fileexchange/67429-a-simple-implementation-of-particle-swarm-optimization-pso-algorithm>
28. A. Askarzadeh, A. Rezazadeh, *Int J Energy Res* **35**, 1258 (2011)
29. A. Askarzadeh, A. Rezazadeh, *International Journal of Electrical Power and Energy Systems* **33**(4), 933 (2011). DOI <https://doi.org/10.1016/j.ijepes.2010.12.036>
30. C. Dai, W. Chen, Z. Cheng, Q. Li, Z. Jiang, J. Jia, *International Journal of Electrical Power and Energy Systems* **33**(3), 369 (2011). DOI <https://doi.org/10.1016/j.ijepes.2010.08.032>
31. A. Askarzadeh, A. Rezazadeh, *International Journal of Hydrogen Energy* **36**(8), 5047 (2011). DOI <https://doi.org/10.1016/j.ijhydene.2011.01.070>
32. J. Zhang, A. Sanderson, *IEEE Transactions on Evolutionary Computation* **13**(45), 945 (2009). DOI 10.1109/TEVC.2009.2014613
33. O.E. Turgut, M.T. Coban, *Ain Shams Engineering Journal* **7**(1), 347 (2016)
34. X. Zhu, N. Wang, *Engineering Applications of Artificial Intelligence* **85**, 740 (2019). DOI <https://doi.org/10.1016/j.engappai.2019.07.019>. URL <http://www.sciencedirect.com/science/article/pii/S0952197619301836>
35. F. Qin, P. Liu, H. Niu, H. Song, N. Yousefi, *Energy Reports* **6**, 1224 (2020). DOI <https://doi.org/10.1016/j.egyr.2020.05.006>. URL <http://www.sciencedirect.com/science/article/pii/S2352484720305199>
36. W. Han, D. Li, D. Yu, H. Ebrahimian, *Energy Sources, Part A: Recovery, Utilization, and Environmental Effects* **0**(0), 1 (2019). DOI 10.1080/15567036.2019.1676845. URL <https://doi.org/10.1080/15567036.2019.1676845>
37. Z. Yuan, W. Wang, H. Wang, A. Yildizbasi, *Energy Reports* **6**, 1106 (2020). DOI <https://doi.org/10.1016/j.egyr.2020.04.032>. URL <http://www.sciencedirect.com/science/article/pii/S2352484720300639>
38. M. Kandidayeni, A. Macias, A. Khalatbarisoltani, L. Boulon, S. Kelouwani, *Energy* **183**, 912 (2019). DOI <https://doi.org/10.1016/j.energy.2019.06.152>. URL <http://www.sciencedirect.com/science/article/pii/S0360544219312848>
39. B. Yang, J. Wang, L. Yu, H. Shu, T. Yu, X. Zhang, W. Yao, L. Sun, *Journal of Cleaner Production* **265**, 121660 (2020). DOI <https://doi.org/10.1016/j.jclepro.2020.121660>. URL <http://www.sciencedirect.com/science/article/pii/S0959652620317078>
40. M. Mansouri, K. Roozrok, F. Jahantigh, *Multiscale and Multidiscip. Model. Exp. and Des* **3**, 1 (2020). DOI <https://doi.org/10.1007/s41939-019-00056-z>
41. A.A. El-Fergany, *Renewable Energy* **119**, 641 (2018). DOI <https://doi.org/10.1016/j.renene.2017.12.051>. URL <http://www.sciencedirect.com/science/article/pii/S0960148117312557>
42. Y. Amadane, H. Mounir, A.E. marjani, E.M. Karim, in *2018 6th International Renewable and Sustainable Energy Conference (IRSEC)* (2018), pp. 1–6
43. J. Shen, J. Zhou, N.G. Astrath, T. Navessin, Z.S.S. Liu, C. Lei, J.H. Rohling, D. Bessarabov, S. Knights, S. Ye, *Journal of Power Sources* **196**(2), 674 (2011). DOI <https://doi.org/10.1016/j.jpowsour.2010.07.086>. URL <https://www.sciencedirect.com/science/article/pii/S0378775310013091>
44. C. Chan, N. Zamel, X. Li, J. Shen, *Electrochimica Acta* **65**, 13 (2012). DOI <https://doi.org/10.1016/j.electacta.2011.12.110>. URL <https://www.sciencedirect.com/science/article/pii/S0013468611019487>
45. G. Pavliotis, A. Stuart, *Springer Science & Business Media* (2008). DOI 10.1007/978-0-387-73829-1

46. M.K. Kadalbajoo, K.C. Patidar, *Applied Mathematics and Computation* **130**(2), 457 (2002). DOI [https://doi.org/10.1016/S0096-3003\(01\)00112-6](https://doi.org/10.1016/S0096-3003(01)00112-6). URL <https://www.sciencedirect.com/science/article/pii/S0096300301001126>
47. J.A. Kierzenka, L.F. Shampine, *Journal of Numerical Analysis, Industrial and Applied Mathematics* **3**(1-2), 27 (2008)
48. W. Wong, C.I. Ming, in *2019 7th International Conference on Smart Computing Communications (ICSCC)* (2019), pp. 1–5
49. K. Priya, T.S. Babu, K. Balasubramanian, K.S. Kumar, N. Rajasekar, *Sustainable Energy technologies and assessments* **12**, 46 (2015)
50. S.I. Valdez, A. Hernández, S. Botello, *New Achievements in Evolutionary Computation*, Peter Korosec (Ed.), ISBN: 978-953-307-053-7, InTech (2010)
51. S. Labs, MATLAB Central File Exchange (2020). URL <https://www.mathworks.com/matlabcentral/fileexchange/74132-genetic-algorithm>
52. X. Meng, Z. Pian, Elsevier pp. 15–50 (2016). DOI <https://doi.org/10.1016/B978-0-12-849896-5.00002-7>
53. J. Kennedy, R. Eberhart, *Proceedings of the IEEE International Conference on Neural Networks Perth, Australia*, 1942–19455 (1995)
54. E. Mezura-Montes, C.C. Coello, *Swarm and Evolutionary Computation* p. 173–194 (2011)
55. M.E. Pedersen, Luxembourg: Hvass Laboratories (2010)
56. D.E. Goldberg, K. Deb, H. Kargupta, G. Harik, In S. Forrest, editor, *Proceedings of the Fifth International Conference on Genetic Algorithms*. Morgan Kaufman **1**, 56 (1993)
57. D.E. Goldberg, B. Korb, K. Deb, *Complex Systems* **3**(5), 493 (1989)
58. S. Bandyopadhyay, H. Kargupta, G. Wang, In *Proceedings of the 1998 IEEE International Conference on Evolutionary Computation*. IEEE Press pp. 603–608 (1998)
59. H. Kargupta, In *Proceedings of the 1996 IEEE International Conference on Evolutionary Computation* pp. 631–636 (1996)
60. H. Kargupta, D.E. Goldberg, In R. W. Belew and M. Vose, editors, *Foundations of Genetic Algorithms 4*. Morgan Kaufmann (1997)
61. F.G. Lobo, K. Deb, D.E. Goldberg, G.R. Harik, L. Wang, In *Genetic Programming 1998: Proceedings of the Third Annual Conference*, Morgan Kaufman pp. 551–558 (1998)
62. C.H.M. van Kemenade, *Proceedings of the 1998 International Conference on Evolutionary Computation*. IEEE Press (1998)
63. H. Mühlenbein, G. Paaß, In *Lecture Notes in Computer Science 1411: Parallel Problem Solving from Nature-PPSN IV* pp. 178–187 (1996)
64. P. Larrañaga, J.A. Lozano, Kluwer Academic Pub. **1** (2002)
65. P. Larrañaga, R. Etxeberria, J.A. Lozano, J.M.P. na, In Boutillier, C. and Goldszmidt, M., editors, *Uncertainty in Artificial Intelligence, UAI-2000* **1**, 343 (2000a)
66. P. Larrañaga, R. Etxeberria, J.A. Lozano, J.M.P. na, In Wu, A. S., editor, *Proc. of the Genetic and Evolutionary Computation Conference, GECCO-2000, Workshop Program* **1**, 201 (2000b)
67. P.A.N. Bosman, D. Thierens, *Proceedings of the Genetic and Evolutionary Computation Conference GECCO-99* **1**, 60 (1999)
68. J.A. Salva, A. Iranzo, F. Rosa, E. Tapia, E. Lopez, F. Isorna, *International Journal of Hydrogen Energy* **41**(43), 19713 (2016). DOI <https://doi.org/10.1016/j.ijhydene.2016.03.136>. URL <http://www.sciencedirect.com/science/article/pii/S0360319915314154>. The 5th Iberian Symposium on Hydrogen, Fuel Cells and Advanced Batteries (HYCELTEC 2015), 5-8 July 2015, Tenerife, Spain
69. E.A. Ticianelli, *Journal of The Electrochemical Society* **135**(9), 2209 (1988). DOI 10.1149/1.2096240
70. Z. Qi, A. Kaufman, *Journal of Power Sources* **113**(1), 37 (2003). DOI [https://doi.org/10.1016/S0378-7753\(02\)00477-9](https://doi.org/10.1016/S0378-7753(02)00477-9). URL <http://www.sciencedirect.com/science/article/pii/S0378775302004779>

A Parameter values for simulations

Table 7 Parameter values used in the baseline calculation used in [5] for dataset 1.

Base parameters	
$N_{cell} = 1$	$A_s = 112 \times 10^4 \text{ (cm g}^{-1}\text{)}$
$R = 8.315 \text{ (JK}^{-1} \text{ mol}^{-1}\text{)}$	$\alpha_a = 0.5$
$F = 96485 \text{ (C mol}^{-1}\text{)}$	$\alpha_c = 1$
$P_{O_2} = 0.21 \text{ (atm)}$	$\%Pt = 0.2$
$T = 308 \text{ (K)}$	$E_{nerts} = 1.23 \text{ (V)}$
$C_{O_2}^{ref} = 1.2 \times 10^{-6} \text{ (mol cm}^{-3}\text{)}$	$\epsilon_{GDL} = 0.3$
$l_c = 1.18 \times 10^{-3} \text{ (cm)}$	$L_{g,c} = 0$
$l_{GDL} = 100.0 \times 10^{-2} \text{ (cm)}$	$\sigma_S = 7.27 \text{ (S cm}^{-1}\text{)}$
$D_{O_2,N} = 1.844 \times 10^{-6} \text{ (cm}^2 \text{ s}^{-1}\text{)}$	$\sigma_N = 0.17 \text{ (S cm}^{-1}\text{)}$
$D_{O_2,W} = 3.032 \times 10^{-5} \text{ (cm}^2 \text{ s}^{-1}\text{)}$	$\rho_C = 2.0 \text{ (g cm}^{-3}\text{)}$
$D_{O_2,GDL} = 2.396 \times 10^{-1} \text{ (cm}^2 \text{ s}^{-1}\text{)}$	$R_{ohm} = 0.47 \text{ (V)}$
$i_0^{ref} = 10^{3.507-4001/T}$	
Initial condition for O_2	
$O_2(0) = \frac{1}{K_{O_2}} \left(\frac{P_{O_2}}{RT} - \frac{I_{\delta} l_{GDL}}{n F D_{O_2,gdl}^{eff}} \right), \quad D_{O_2,gdl}^{eff} = D_{O_2,GDL} \epsilon_{GDL}^{\frac{3}{2}}$	
$K_{O_2} = \frac{1}{RT} \exp \left(-\frac{666}{T} + 14.1 \right)$	
Reference values to estimate	
$m_{Pt} = 0.332 \text{ (mg cm}^{-2}\text{)}$	$\rho_{Pt} = 21.5 \text{ (g cm}^{-3}\text{)}$
$\epsilon_c = 0.33$	

Table 8 Parameter values used in the baseline calculation used in [6] for dataset 2.

Base parameters	
$N_{cell} = 1$	$R_{ohm} = 0.225 \text{ (V)}$
$R = 8.315 \text{ (JK}^{-1} \text{ mol}^{-1}\text{)}$	$\alpha_a = 0.5$
$F = 96485 \text{ (C mol}^{-1}\text{)}$	$\alpha_c = 1$
$P = 5 \text{ (atm)}$	$\sigma_N = 0.17 \text{ (S cm}^{-1}\text{)}$
$T = 353.15 \text{ (K)}$	$\sigma_S = 7.27 \text{ (S cm}^{-1}\text{)}$
$C_{O_2}^{ref} = 1.2 \times 10^{-6} \text{ (mol cm}^{-3}\text{)}$	$x_{O_2} = 0.21$
$l_c = 10 \text{ (}\mu\text{m)}$	$L_{g,c} = 0.1$
$l_{GDL} = -$	$L_{m,c} = 0.4$
$D_{O_2,W} = 9.19 \times 10^{-9} \text{ (m}^2 \text{ s}^{-1}\text{)}$	$\rho_C = 1800 \text{ (kg m}^{-3}\text{)}$
$m_C = 4.5 \text{ (mg cm}^{-2}\text{)}$	$E_{nerts} = \text{Eq. (8)}$
Expressions in Table 2	
f, A_s, i_0^{ref}	
Reference values to estimate	
$m_{Pt} = 0.35 \text{ (mg cm}^{-2}\text{)}$	$\rho_{Pt} = 21.4 \text{ (g cm}^{-3}\text{)}$
$\epsilon_c = 0.4$	

Table 9 Parameter values used in this work to adjust dataset 3 obtained from [68].

Base parameters	
$N_{cell} = 1$	$R_{ohm} = 0.84225$ (V)
$R = 8.315$ (JK ⁻¹ mol ⁻¹)	$\alpha_a = 1$
$F = 96485$ (C mol ⁻¹)	$\alpha_c = 1$
$P = 1.6$ (atm)	$\sigma_N = 0.17$ (S cm ⁻¹)
$T = 353.15$ (K)	$\sigma_S = 7.27$ (S cm ⁻¹)
$C_{O_2}^{ref} = 1.2 \times 10^{-6}$ (mol cm ⁻³)	$x_{O_2} = 1.0$
$l_c = 12$ (μ m)	$L_{g,c} = 0.1$
$l_{GDL} = -$	$L_{m,c} = 0.01$
$D_{O_2,W} = 0.7 \times 10^{-8}$ (m ² s ⁻¹)	$\rho_C = 1800$ (kg m ⁻³)
$m_C = 4.5$ (mg cm ⁻²)	$E_{nerts} = \text{Eq. (8)}$
Expressions in Table 2	
f, A_s, i_0^{ref}	
Parameters to estimate	
m_{Pt} (mg cm ⁻²), m_C (mg cm ⁻²), ϵ_c	