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Proposal of Multidisciplinary Design Optimization Approach for a Proton Exchange Membrane Fuel Cell Performance Estimate

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Abstract: - The amount of current density the stack is able to produce is the key performance indicator for a fuel cell system; at given chemical conditions and geometry, the leading parameters in a Proton Exchange Membrane fuel cell behavior are the exchange current density, both at anode and cathode, as well as the temperature and relative humidity at the anode. Such considerations lead to the definition of a surrogate model that is subsequently validated. Such model is then used as the basis for a Multidisciplinary Design Optimization process based. A comparison among suitable approximation methods is considered with the aim to reduce the computational time. The presented work focuses on two unconstrained single-objective optimization processes to find the best solution in terms of maximum current density produced at a given voltage. Finally, the optimized outputs are validated.

Key-Words: - PEM Fuel Cell; Multidisciplinary Design Optimization; Sensitivity Analysis; Monte Carlo Simulation; Surrogate Model

1 Introduction

New research areas are continuously being investigated in the effort to reduce environmentally harmful emissions. The final objective of the presented environmental research is to provide an innovative and advanced methodology able to help researchers pursuing the reduction of harmful emissions. Proton Exchange Membrane Fuel Cell (PEMFC) technology is increasingly gaining interest in the field of movable energy sources with reduced environmental impact. The improvement of computational capabilities enables to study models with growing complexity, allowing a better comprehension of fuel cell behaviour during the design phase.

PEMFCs typically uses a water-based, acidic polymer membrane as its electrolyte, with platinum-based electrodes that split the hydrogen into positive ions (H^+ , protons) and negative electrons. H^+ ions pass through the membrane to the cathode to combine with oxygen to produce water. Electrons must pass round an external circuit creating a current to rejoin H^+ ions on the cathode. PEMFCs operate at relatively low temperatures (typically below $100\text{ }^{\circ}\text{C}$) and can tailor electrical output to meet dynamic power requirements; due to the relatively low temperatures and the use of precious

metal-based electrodes, these cells must operate on pure hydrogen. Oxygen can be provided in a purified form, or directly extracted at the electrode from atmospheric air.

The design phase of a fuel cell system is extremely important and delicate, as all its features must be correctly identified to comply with the imposed requirements. Therefore, the definition of a suitable design strategy gets crucial. One of the main possible issues consists in adequately managing any design change that could become necessary in a less time consuming manner, while also pointing at setting up a robust and confident simulation framework.

The goal of this paper is to outline the best suitable approach to perform a Multidisciplinary Design Optimization (MDO) process of a PEM fuel cell, starting from its distributed parameters model and resulting in a surrogate model, predicting the cell behaviour, as explained in the following paragraphs. The sensitivity analysis performed in [1] is considered as the basis to the construction of the surrogate model to be optimized (also referred to [2, 3]). A review of methods dedicated to numerical optimization processes is provided by Secannell et al. [4]; the approach provided in Mukhtar et al. [5] is considered as particularly relevant.

The presented work combines a Computational Fluid Dynamics (CFD) model, a Design of Experiment (DoE), surrogate models and optimization algorithms in an automated way with the aim of obtaining a complete optimization loop for PEM fuel cells. The optimization process of fuel cell performances relates to the design of an innovative hydrogen-fueled electrical glider, with the complete power generation and management is being designed at the Department of Mechanical and Aerospace Engineering (DIMEAS) of Politecnico di Torino.

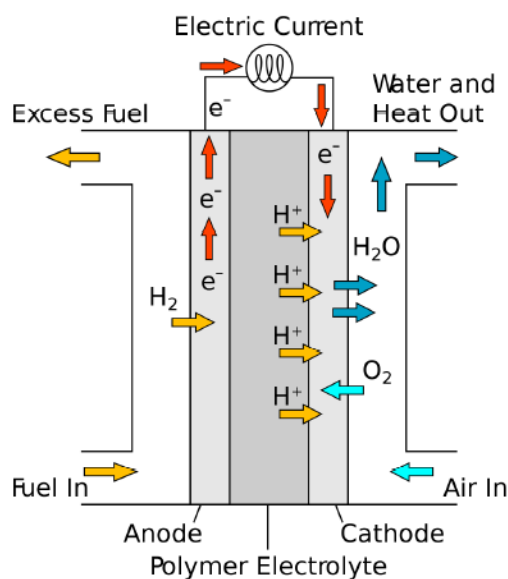


Fig. 1: Schematic representation of a Proton Exchange Membrane Fuel Cell (PEMFC); it is possible to see some of the components and key factors affecting FC performance and behavior, as the hydrogen (fuel) flow and the oxygen (air) flow, both with their respective temperature and humidity; protons flow through the membrane, from anode to cathode;(origin: Wikimedia).

2 Methodology

The design of an optimized PEMFC is a complex process that involves several synergic activities.

Starting from the preceding considerations, the main structure of the presented work can be summarized with the following steps:

1. Setup of the PEM fuel cell CFD model.
2. Design space evaluation
3. Surrogate model creation
4. Surrogate model-based optimization
5. Validation

Starting from [1], steps 1 and 2 are covered by an assessment of available PEMFC simulation models available in the literature and by a sensitivity analysis able to individuate the key parameters to be considered in surrogate modelling.

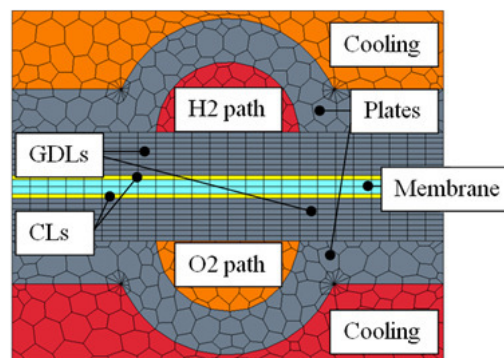


Fig. 2: model used to represent the fuel cell section.

2.1 Model setup

The PEM fuel cell model implemented for this work is the same discussed and validated in [6] and also used in [1]. This model simulates a small portion of a single fuel cell area (3 mm x 10 mm), but considering all of the fluidic and solid components of a FC. The elements modelled are visible in Fig. 2.

Their main fluid-dynamics characteristics are:

- Three-dimensional model,
- Steady state,
- Simple, mono-dimensional, complete electric field,
- Non isotherm,
- Multi-gas component but with consideration of liquid water effects, and
- Structural and anisotropic thermal properties.

Gaseous species consumption and production are implemented as sources / sinks in the mass conservation equations. On the other hand, from an electro-chemical point of view, the model implemented considered all the main aspects involved in a fuel cell operation, i.e.:

- Ideal voltage (Nernst voltage considering pressure-increased voltage),
- Ohmic voltage losses (due to the electric resistance of the PEM membrane),
- Electrochemical activation voltage losses (due to electrochemistry), and
- Concentration losses (due to the finite gaseous species diffusion over the catalytic surfaces).

Due to the steady-state nature of the model, capacitive and inductive phenomena were not modelled, despite their strong influence during transient PEMFC operation. The resulting electrical model can be simply represented with series resistances (Fig.3), where the cell voltage is assumed constant over the cell surface, while the electric current, as well as resistances and ideal voltage is the unknown variable calculated point by point over the cell surface and as a function of the local operating parameters.

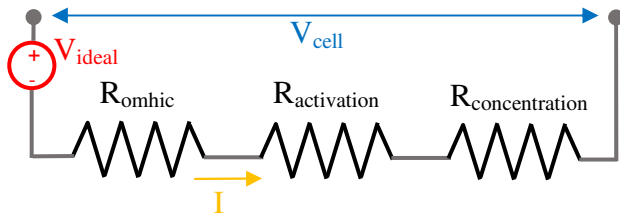


Fig. 3: Equivalent electrical model of the PEMFC CFD model. Ohmic (R_{ohmic}) losses, activation ($R_{activation}$) and concentration ($R_{concentration}$) losses were considered.

The ideal voltage (V_{ideal}) is calculated as function of reactants pressure and cell temperature. Cell voltage (V_{cell}) is superimposed as boundary condition. Electric current (I) is the unknown variable. All of these variables (except V_{cell}) are calculated point by point over the entire cell area. The three main voltage losses modelled as resistances also give the shape of the typical fuel cell polarization curve (voltage vs. current density in A/cm^2) shown in Fig. 4 and obtained with the FC model here adopted.

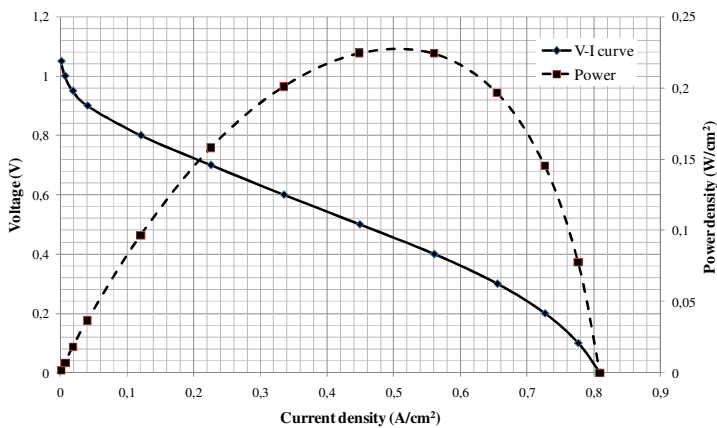


Fig. 4: Fuel cell polarization curve obtained with the CFD model here used. Both Voltage (V) and power density (W/cm^2) vs. current density (A/cm^2) are shown.

The main voltage losses modelled are visible in the voltage curve of Fig. 3: initial and final voltage curved drops are given by activation and concentration losses, respectively, while the straight portion and its steepness is given by ohmic losses.

2.2 Design space evaluation

The sensitivity analysis has been based on a numerical simulation model correlated with experimental data. Several reviews of PEMFC models have been considered from available literature [4-11]. Two sets of parameters (design variables) have been identified:

- **Noise factors:** these are the boundary conditions values, also defined uncontrollable input noises;
- **Control factors:** these are the tuning parameters or controllable inputs.

The cross-correlation between noise and control factors proven to be low. Therefore, a sensitivity analysis has been performed to assess the impact of each parameter on the current density (that has been considered as the key performance indicator).

Exchange current density both at **anode** and **cathode** are the leading noise factors; **temperature and relative humidity at the anode** are the most impacting control factors; geometrical parameters are also important for PEMFC behaviour, but the optimization considered in this paper is given for fixed fuel cell geometry. As a consequence, the surrogate modelling has been completed considering these factors as the leading parameters. In detail, these parameters are described in the following.

Cathode exchange current density, i_{0c} : the exchange current density is an important electrochemical parameter related to the kinetics of the chemical reactions. This variable depends upon many physical and electro-chemical factors, as the noble metal particles used, their shape and distribution over the catalytic surfaces and their micro-structure; in the model, it is defined for both the cathode and the anode sides. This variable is usually measured in A/cm^2 . The higher its value, the faster the chemical reactions. A quicker chemical reaction has the direct effect of lowering the detrimental voltage losses, since it implies a lower amount of energy absorbed by the reaction itself (as a voltage loss), improving the power output [12].

Anode exchange current density, i_{0a} .

Anode inlet gas temperature, T_a : the temperature of the gas mixture entering at the hydrogen side [13].

Anode inlet relative humidity, Rh_a : the relative humidity of the gas mixture entering the cell at the hydrogen side. In case of PEMFCs, the polymer membrane requires high level of humidity to operate properly as electrolytic element of the cell [14].

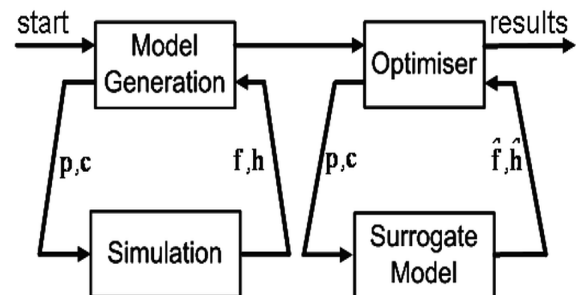


Fig. 5: Logical scheme of the sequential approach used in this study. The “model generation” and the “simulation” blocks refers to the “real model” on which the surrogate model is built for the optimization. The sensitivity analysis and the corresponding DoE are performed at the “simulation” block level [1].

Statistics is extensively used at the beginning of this methodology, varying the model inputs to correctly estimate the system output changes through a sensitivity analysis. Furthermore, a DoE approach is used to create a design matrix required to provide the anchor points of the approximation model, correlating design inputs and objective functions.

This approach, better known as Response Surface Methodology (RSM) is extensively adopted [1] to speed up the following MDO process. A Monte Carlo Simulation (MCS) is chosen to evaluate how key parameters affect the final results [15]. The selected variables are sampled through a certain distribution (described later) and a sensitivity analysis is performed. Graphical methods can be adopted to better understand the results [16]. After evaluating the computational time of a single run of the multidisciplinary PEM fuel cell model, the authors decided to build up a surrogate model, according to literature [16, 17]. In this paper, some available approximation algorithms are investigated and compared with the aim of selecting the most suitable one, on the basis of the minimum error percentage [17]. As a final point, two unconstrained deterministic single-objective MDO processes are performed to identify the best set of parameters to produce the maximum current density, i.e. the maximum output power. These analyses are obtained recurring to an evolutionary technique, more suitable to find the global optimum than a gradient-based algorithm [16]. A commercial tool (*iSight*) is chosen to set up the whole process, embedding the multidisciplinary fuel cell model (involving CFD and electrochemical codes), the DoE, the surrogate model and the single-objective optimization [18]. The complete process performed in this work is shown in Fig. 5 [1].

2.3 Surrogate modelling

Many engineering analyses consist in running complex computer codes, requiring a vector of design variables x (inputs) and computing a vector of responses y (outputs). Despite the significant technology advances in the information technology field, the expense of running finite elements analyses is still sensible, as they can take minutes to hours, or even longer, to be completed. Moreover, this query-and-response technique often leads to a trial and error approach, where the designer will almost never find out the functional relationship between x and y . This means the best settings for the input values will be very difficult to be identified [3, 19, 20]. Statistics-based techniques are widely used in engineering design to address these concerns.

The basic approach is to build approximated models of the analysis codes, able to get the results in a shorter time.

If the true behaviour of a computer analysis code is shown as

$$y = f(x) \quad (1)$$

then its meta-model can be represented as

$$\hat{y} = g(x) \quad (2)$$

with

$$y = \hat{y} + \varepsilon \quad (3)$$

where ε stands for the approximation errors and \hat{y} is the meta-model. These methods are extremely useful to reduce the computational time. However, it is fundamental to guarantee the goodness of the solution. Modern optimization methods and especially global optimization approaches could lead to macroscopic mistakes if not adequately managed. For this reason, a significant number of design evaluations are required to build meta-models able to simulate the real nature of approximated codes. Surrogate models are simplified, analytical approximations, based on few supporting points obtained from the simulations of the original evaluation model. Due to their simplicity they allow a low cost prediction of the system behaviour with an arbitrary elevated number of design evaluations during the optimization loop [19]. At the same time, their simplicity could lead to numerical or actual errors, due to the fact that approximated functions could miss the real shape of the domain. Validation and refinements of approximated models are then necessary before continuing with next optimization steps [20, 21].

In this work, four main techniques are evaluated to find the most suitable one. The algorithms taken into account in this work are the RSM, the Radial Basis Function (RBF), the Kriging and the Chebyshev/Orthogonal Polynomial. An advantage of using orthogonal functions as a basis for fitting is that the inputs can be decoupled in the analysis of variance [22]. Chebyshev orthogonal polynomials are a common type of orthogonal polynomials that are particularly useful for equally spaced sample points. They are used when the sampling strategy is an orthogonal array. The *iSight* software used for this analysis implements Taguchi's method [23] for fitting Chebyshev polynomials from an orthogonal array. A minimum number of samples have to be produced to obtain a surrogate model. According to Fig. 1, the analytical model has to be run with the aim of generating the needed points.

According to the Taguchi's theory, the model generation was obtained thanks to two sensitivity analyses of both noise factors and control factors [2, 24]. The collected data were used to build up the approximated models.

2.4 Optimization process and validation

An exploratory technique was used to better investigate the design space. An optimization process is very sensible to the chosen optimization strategy. A gradient-based algorithm usually provides a local optimum, without evaluating any other feasible optima if the design space is not linearly defined. The choice of an evolutionary technique (e.g. a genetic algorithm GA) is motivated by the desire to investigate the whole design space, with the scope of finding the global optimum [16, 25, 26], avoiding to get stuck in local solutions. Several genetic algorithms are available in literature. In this paper the Multi-Island GA (MIGA) was used [27, 28]. This technique can be implemented even if the optimization problem is not a multi-objective one. Inside a MIGA, a design point is named an individual, and is associated with a value of fitness, obtained from the value of the objective function and constraint penalty. Better values of the objective function and low penalty rates build up a higher fitness value. The peculiarity of MIGA is that the overall population of individuals is not unique, as design points are grouped into several sub-populations, called islands. The typical iterations of genetic algorithms are carried out independently within each island. Individuals are selected and moved from one island to another one in each iteration, in what is called a migration, that is regulated by the migration interval (number of generations between each migration) and the migration size (the percentage of individuals in the population that are moving at the time of migration) [5, 18, 26]. The deterministic single-objective optimization consisted in maximizing the current density at a given voltage, without imposing any constraint. The optimization performed was a deterministic one, according to the previous generated data. The approximated model was built to speed up the optimization process. The results were carried out at a voltage of 0.2 V, since this area of the polarization curve is characterized by possible flooding phenomena and by a sharp decrease in the generated power, hence making the model more sensible to changes. Therefore, it can be assumed that the validation of the surrogate model in such a condition extends the applicability of the same approach at any other voltage within the cell operating range.

3 Results and discussions

3.1 Surrogate modelling and error estimate

Two tables were obtained, summing up the error analysis. The first presented case study is the approximation of the control factors matrix. The other set of parameters, the noise factors, were set as constant: their values are shown in Table 1.

Four approximation techniques were investigated to evaluate which one is the most performing. A useful tool to understand the quality of the surrogate model is the error analysis. To perform an error analysis [29], some points are requested for a cross-validation: a number of data points were removed from the sampling data set, one at a time. For each of the removed points, the approximation coefficients were re-calculated, and the exact and approximate output values were compared. The removed point was then put back into the data set and the next point was removed. The choice of points is random, and the total amount of the points was equal to the number of points generated by the DoE, performing in this way a more detailed error analysis. As shown in Table 2, the Kriging approximation method provides an average and maximum error about three times the error obtained with other methods. Moreover, the RBF technique is the most performing (1.816% of average error) and the orthogonal polynomial and RSM approximation also provide an acceptable error ($\sim 2.7\%$ of average error). The R^2 value, also known as the coefficient of determination, is the ratio of the explained variation to the total variation. It is a mathematical measure of the error which estimates with a single number how well the assumed functional form of the response measures the variability of the supplied response data. A perfect fit of the response data corresponds to an R^2 value of 1.0. R^2 is defined as:

$$R^2 = \frac{\sum(\hat{y}_i - \bar{y})^2}{\sum(y_i - \bar{y})^2} \quad (4)$$

where: y_i is the observed value for the i^{th} row of the DoE, \hat{y}_i is the predicted value for the i^{th} row of the DoE and \bar{y} is the average of the observed values [29].

r_{cond} (1/s)	i_{0a} (A/m ²)	i_{0c} (A/m ²)	sat_{rate} (-)
200	1700	2	400

Table 1: Constant values used for the noise factors when building the surrogate model for the control factors.

<i>Approximation technique</i>	<i>Technique options</i>	<i>Number of cross-validation points (-)</i>	<i>Average error (%)</i>	<i>Maximum error (%)</i>	<i>R² (-)</i>
RSM	Quadratic	99	2.727	15.429	0.91647
RBF	-	99	1.816	14.604	0.92013
Orthogonal	Chebyshev 3° order polynomial	99	2.628	15.713	0.91542
Kriging	-	99	6.072	44.231	0.70623

Table 2: Approximation techniques used to build the surrogate models for the control factors. The table shows the average and maximum error committed using the corresponding surrogate model. Also R^2 values are reported.

Kriging approximation method provides an R^2 value that is quite far from the unit value if compared to the results provided from the other methods, showing the worst approximation. Fig. 6 provides a useful plot to give a better idea of the entity of the average error obtained thanks to the RBF. It is an actual vs. predicted plot showing the actual values of the response (obtained with the CFD analysis) plotted against the predicted equation for the response based on the assumed functional form. An even distribution of the data along the perfect fit line (in black) indicates smoothness of the assumed model and provides an overview of the shape of the model error [29]. As a conclusion, the RBF method was chosen for the next optimization process. Nevertheless, a validation process is needed to verify if the point is a real physical value. These analyses will follow the optimization process and are presented in the following. As done before, the same approximation techniques were investigated also for the noise factors, keeping constant the control factors values (as shown in Table 3).

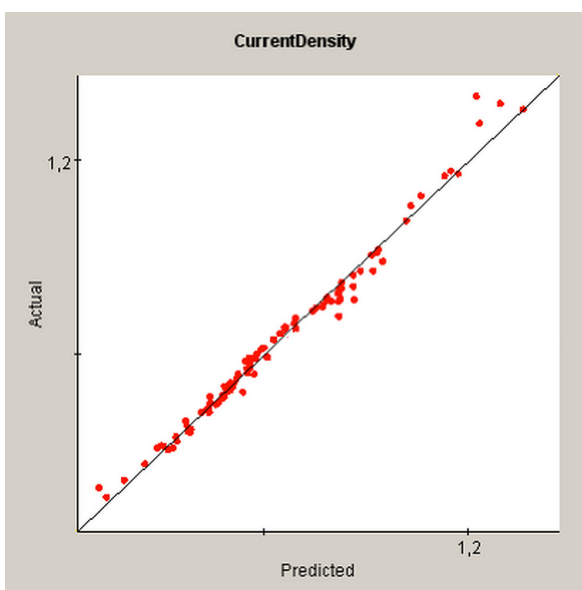


Fig. 6: Plot of the error given by the RBF approximation.

<i>Rh_a</i> (%)	<i>T_a</i> (K)	<i>Rh_c</i> (%)	<i>T_c</i> (K)	<i>Compr</i> (%)
90	300	85	310	15

Table 3: Constant values used for the control factors when building the surrogate model for the noise factors.

The error analysis is presented in Table 4, providing a useful decision tool. The Kriging approximation method provides an average error that is about two times the error obtained with the RSM and orthogonal method, while it is about three times the error given by the RBF technique. Moreover, the Kriging method provides a maximum error that is greater than the errors obtained with the other methods. The other techniques, instead, provide very similar results. The Kriging approximation method provides an R^2 value that is quite far from the unit value if compared to the results provided from the other methods, showing the worst approximation. Even if the orthogonal approach presents some better performance if compared with the RBF methodology, the latter was tested to verify if it could be used to create also this surrogate model. Other error analysis tools could be used, e.g. the Model Fit Error (MFE) and the Model Representation Error (MRE) [20].

3.2 Optimization process and validation

At this point, two different optimization processes were performed, depending on which set of parameters was fixed. The results are presented for two different sets, one set managing the noise factors values and a second one using the control factors values. As outlined in the previous chapter, it should be noted that the optimizations were performed on the approximate models, obtained thanks to the RBF technique. As reported in Table 5 and 6, for the genetic algorithm some population parameters were set: 20 subpopulations were created for each one of the 20 different islands, defining a sufficient number of samples (400).

<i>Approximation technique</i>	<i>Technique options</i>	<i>Number of cross-validation points (-)</i>	<i>Average error (%)</i>	<i>Maximum error (%)</i>	<i>R² (-)</i>
RSM	Quadratic	99	3.427	37.830	0.90852
RBF	-	99	2.582	34.576	0.90468
Orthogonal	Chebyshev 3° order polynomial	99	3.299	32.115	0.91932
Kriging	-	99	5.819	52.143	0.67478

Table 4: Approximation techniques used to build the surrogate models for the noise factors. The table shows the average and maximum error committed using the corresponding surrogate model. Also R2 values are reported.

<i>Approximation technique</i>	<i>Optimization algorithm</i>	<i>Optimized noise factors</i>				
		<i>r_{cond} (1/s)</i>	<i>i_{0a} (A/m²)</i>	<i>i_{0c} (A/m²)</i>	<i>sat_{rate}</i>	<i>Current density (A/m²)</i>
RBF	Multi-Island 20 subpopulations, 20 islands	-153.967	2999.762	4.9994	632.082	0.7614

Table 5: Optimization results for the noise factors values. In the table are reported both the optimal noise factors values and the corresponding optimized objective function (current density).

<i>Approximation technique</i>	<i>Optimization algorithm</i>	<i>Optimized control factors</i>					
		<i>Rh_a (%)</i>	<i>T_a (K)</i>	<i>Rh_c (%)</i>	<i>T_c (K)</i>	<i>compr (%)</i>	<i>Current density (A/cm²)</i>
RBF	Multi-Island 20 subpopulations, 20 islands	0.9991	335	0.9917	300	0.335	1.7266

Table 6: Optimization results for the control factors values. In the table are reported both the optimal control factors values and the corresponding optimized objective function (current density).

A very important result was obtained: the optimized current density value is about 0.76 A/cm² in the case of the noise factors optimization, i.e. keeping the control factors constant. The baseline starting current density value was 0.72 A/cm² at a voltage of 0.2 V. It must be underlined that noise factors are not controllable and modifiable, since their values depend mainly on physical laws.

The second optimization uses the control factors as design variables, while the noise factors are set constant. The results are given in Table 6. As shown, the same number of population parameters was set. Compared to the previous optimization loop, a different result was obtained: the optimized current density value is about 1.7266 A/cm², much higher than the starting value.

The control factors can be tuned easily by the user if compared to the noise factors, making them the real key to optimize fuel cell operations. A validation process is needed to verify if optimal solutions found by the genetic algorithm is physically acceptable [30] and in accordance with a direct CFD simulation.

A validation was done recurring to the multidisciplinary CFD cell model, to extract the simulated value of current density – the predicted "real" value, and compare it to the one estimated by the surrogated approach. The results, reported in Table 7, are extremely accurate for both the approximated optimization processes, confirming the goodness of the surrogate modelling technique and validating it completely. All of the simulations were obtained with an HP xw6600 Workstation equipped with Intel Xeon architecture, with four dual core units (eight processors) and 8 GB of RAM. The average time required for obtaining a single operating point of the fuel cell polarization curve using a complete CFD simulation is around 15 minutes, considering the fluid dynamics solution sufficiently converged. The total time required to obtain the sensitivity analysis with 100 points (requiring the execution of 100 different CFD simulations), together with the data management and post-processing operations, was of about 3 days and 23 hours. This can be considered the time spent to obtain the surrogate model.

Nevertheless, having such meta-model available, it is possible to obtain a new polarization curve point in the order of few seconds of computational time, instead of using a complete CFD simulation. This allows for a rapid optimization analysis (requiring sometimes hundreds of simulations), requiring about 10 minutes for the whole meta-model-based MDO process.

	<i>Noise factors case study</i>	<i>Control factors case study</i>
Estimated optimal current density (surrogate model) (A/cm ²)	0.7614	1.7266
Simulated current density (CFD model) (A/cm ²)	0.7620	1.7412
Error (%)	0.078	0.839

Table 7: Evaluation of the error committed using the surrogate model for the optimization process.

4 Conclusions

The main objective of this work was to set up an optimization environment dedicated to the design of PEM fuel cells, and to assess its reliability and feasibility when applied to such technologies. This methodology was applied to a relatively simple and small PEM fuel cell model. This work showed the potential of the surrogate modelling technique combined with an optimization process. The main aspects considered involved the setup of an automated MDO process based on a surrogate model obtained thanks to a previous sensitivity analysis performed recurring to a Monte Carlo Simulation. This approach was chosen with the aim of estimating the parameters affection on the global cell performance. The unconstrained deterministic single-objective MDO processes performed, finding the optimal solution in terms of maximum current density the cell can produce at a given voltage, was followed by a validation process of the optimal solution that was tested to confirm the effectiveness and quality of the whole process implemented (i.e. keeping the average error to a minimum value). As a conclusion, this work showed the potentialities of the application of such techniques for the optimal design of fuel cells.

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Abbreviations:

CFD	Computational Fluid Dynamics
CL	Catalyst Layer
DoE	Design of Experiment
GDL	Gas Diffusion Layer
MCS	Monte Carlo Simulation
MDO	Multidisciplinary Design Optimization
MFE	Model Fit Error
MIGA	Multi-Island Genetic Algorithm
MRE	Model Representation Error
PEM	Proton Exchange Membrane
PEMFC	Proton Exchange Membrane Fuel Cell
RBF	Radial Basis Function
RSM	Response Surface Methodology