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Targeting optimized and robust operating conditions in a hydrogen-fed Proton Exchange Membrane Fuel Cell

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ABSTRACT

Response Surface Methodology (RSM) when combined with the Propagation of Error (PoE) approach offers an efficient robust design able to find the best operating conditions to simultaneously maximize power density and reduce normal operation variability in a hydrogen-fed Proton Exchange Membrane Fuel Cell (PEMFC). To proceed with the statistical analysis, a central composite design with 20 experimental runs (6 central points were used to assess the experimental error) was adopted to inspect which factors have significant effects and how they interact each other. This allowed generating a polynomial function to determine the maximum power density at 1415 mW/cm^2. Taking advantage of the desirability concept and using the PoE measure as a response, a multiple optimization under different restrictions was carried out defining a new set of operating conditions able to target the maximum possible power density at the most robust conditions (1074 mW·cm^{-2} at 55 °C, 50% RHC and 25 Psi). Then, actions were carried out to narrow even more the tolerance intervals towards more ambitious standards. Reducing the standard deviation from input factors through the use of adequate controlling measures led to a decrease of almost 50% in the tolerance intervals. This is an useful methodology to help the PEMFC normal operation more repeatable and predictable under its lifetime by combing both optimization and robustness goals.

1. Introduction

Among several types of fuel cells, the Proton exchange membrane fuel cell (PEMFC) is one of the most promising power sources for different scenarios and usages [1–3]. To further optimize PEMFC's efficiency and to increase its power, a thorough understanding is required of factors that influence output performance [4,5]. Indeed, PEMFCs present a complex non-linear multivariate nature where several factors influence significantly the power output. Traditional one by one experiment optimization implies the testing of factors one at a time instead of conducting all of them simultaneously. This approach presents several drawbacks, namely, requiring an excessive number of experiments, missing the optimal set of factors and neglecting the interactions between factors. These interactions could play a key role on the system performance. This suggests the design of experiments approach rather than fundamental or mechanistic models [4–10]. Silva et al. [4] evaluated the power density of a Direct Methanol Fuel Cell (DMFC) as a function of temperature, methanol concentration, air flow rate, methanol flow rate and air relative humidity by using a design of experiments coupled with a Response Surface Methodology (RSM). Authors also applied both methodologies to select the best proton exchange membrane considering different thicknesses. Kanani et al. [6] optimized the cathode stoichiometry, anode stoichiometry, gases inlet temperature, and cathode relative humidity in a serpentine PEMFC using RSM. Results predicted an optimum value for all the operational parameters to reach the best performance. Prediction were validated by experimental runs and appeared to be in good agreement with each other. San et al. [7] used RSM to inspect the effects of roughness of polymer composite bipolar plate, contact angle and hydrogen flow rate on power density of PEMFCs. Optimal values of the contact angle, hydrogen flow rate and roughness were found as 81.2°, 1.87 dm^3 min^{-1} and 1.69 mm, respectively.

Parameter designs (two-array) made popular by Taguchi are other suitable option in order to find optimal operating conditions for quality improvement purposes in different industrial processes [11–15]. Chang et al. [11] combined a generic numerical model with the Taguchi

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method to find the optimal performance of a PEMFC. Their objective was to reduce fuel consumption and operation costs by determining the right parameters for the cell. Results were later benchmarked against experimental data and showed that the combination between Taguchi and numerical model was able to successfully predict the optimal operating point. Wu and Ku [12] analyzed cell performance using rectangular cylinders using a three-dimensional PEMFC model in conjunction with the Taguchi method. Optimal combination of factors were found and results were in line with experimental data.

Although the approach from Taguchi became extreme popular as an effective tool for quality improvement in the 1980s, a large controversy arose because there were significant issues with the advocated experimental strategy and data analysis procedures [16–18]. Additionally, it was concluded that fractional designs deliver considerable more information becoming much more efficient than the two-array parameter designs developed by Taguchi [18]. These major drawbacks can be overtaken by using an advanced approach to robust design making use of RSM [17–18].

Beyond obtaining optimal operating conditions, manufacturers expect to produce fuel cell devices generating robust responses with minimal variations to guarantee reliable performances. Anderson and Whitcomb [19] suggest the addition of propagation of error (POE) as a criterion to find operating conditions robust enough to variation transmitted from input variables and that can be used in a large spectrum of processes. PoE can be defined as the square root of the variance of a selected response. When combined with proper fractional designs, this methodology is more efficient (reduced number of runs) and provides more information than designs advocated by Taguchi [20–22].

The robust analysis of a process combined with an optimization procedure to find the most efficient combination of process variables that might be used during normal operation is missing in the literature. Silva and Rouboa [21] combined RSM with the propagation of error approach to evaluate the potential of Portuguese biomasses to produce syngas for different applications. From their analysis, authors were able to correctly predict the optimal application for several substrates while verifying that the operating parameters necessary to get higher performances did not always match with those necessary to obtain a stable syngas composition, a crucial point that could only be concluded by using advance analysis tools like RSM and PoE.

To cover this significant gap in the current literature an advanced approach to robust design making use of RSM is presented. To do so, a combination between RSM and PoE is performed to determine the most robust operating conditions to achieve maximum power generation from a PEMFC. Based on collected experimental data, a central composite design was selected to determine the best operating conditions using power density output as response. Later, a multi-stage optimization including the PoE as a response to identify the operating conditions less sensitive to variations was conducted. Finally, the system capability was improved by narrowing tolerance intervals and best options were determined to move results towards six sigma standards.

2. Experimental procedures

2.1. Membrane electrode assembly (MEA) fabrication

Anode and cathode catalyst ink were prepared by directly mixing Pt/C catalyst (20 wt% Pt, BASF) with Nafion solution (5 wt% solution, EW1000, Aldrich). Nafion content within the catalyst ink was 30 wt%. The catalyst ink was sonicated for 3 min with isopropyl alcohol as the solvent, and coated onto a gas diffusion medium consisting carbon paper (TGP-0120ST, Fuel Cell store) with microporous layer by the loading of 1.98 $mg \cdot cm^{-2}$. The platinum loadings on both the anode and cathode were 0.38 $mg \cdot cm^{-2}$. Prepared electrodes and membrane 212 were sandwiched to form the MEA with an active area of 5 cm^2. Further details can be found in [23–24].

Table 1
Selected operating conditions and corresponding ranges used for the design of experiments.

Operating conditions	Operating range
Cell temperature, °C	55–75
Cathode (RH_C), %	50–100
Gases pressure, Psi	5–25

2.2. MEA testing

The MEA was subjected to constant voltage 0.6 V for 9 h. During this period, the applied operation condition was: cell temperature of 55 °C, operating pressure of 34 kPa and fully humidified hydrogen and oxygen with flow rate 200 $ml \cdot min^{-1}$. Selected conditions were considered as the most favorable from previous studies [23,24]. MEAs conditioning was interrupted every hour to assess the i-V curves history.

The steady-state performance of activated MEA was evaluated by polarization curves under different operation conditions. Polarization curves were obtained by scanning of the cell voltage from Open circuit Voltage (OCV) to 250 mV, with a scan rate of 5 $mV \cdot s^{-1}$.

2.3. Design of experiments

Standard designs such as Central Composite Design are especially suitable to inspect experimental studies where factors should be bounded within previous known values. Based on previous experiments [23,24], cell temperature, cathode relative humidity (RH_c) and operating pressure ranges were selected as shown in Table 1. The RH of anode was 100% in all temperatures.

Twenty runs were performed changing the factors within the corresponding ranges as shown in Table 2. To get any sort of power to evaluate pure error at least four center points must be replicated [25]. In this work and to get uniform precision six points were included.

3. Discussion

3.1. Statistical model validation

One of the first steps to generate a good predictive statistical model based on the design of experiments methodology is to determine how far it is worth going in the polynomial degree. Indeed, high level terms should be only included if they could explain considerable variation related to the process. One effective method to select the necessary and sufficient terms is the sequential mode sum of squares (SMSS). SMSS shows how much of the remaining variation is explained by including a factor given that other factors are already in the model. Table 3 reports the SMSS analysis for power density as well as F and p-values.

F value is a simple ratio between the variation between sample mean and variation within the samples. For practical purposes and applied to this case, high values of F mean a more significant model. The large F value for the cubic model is misleading because the selected design lacks the necessary points to fit all required terms. This model could be classified as aliased and is not a valuable option. Considering the remaining options and because the quadratic model besides including linear and interaction terms (AB, BC, etc.) also presents a p-value significantly lower than 0.1, it should be selected as the right empirical model to fit the power density within the design space.

Lack-of-fit tests are intimately related with pure error. They present a relationship between the variation of the replicates and variation of the design points about their predicted values. A F representation for the lack-of-fit test can be as follows: