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Influences of Gas Pressures and Flow Rates on the Maximal Power of SOFC by using the Design of Experiment Methodology

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Abstract: ***Different types of fuel cells exist; they transform the chemical energy contained in hydrogen into electrical energy and heat. The solid oxide fuel cell (SOFC) debit the largest electric power due to solid electrolyte which allows the cell to function at very high temperature, thus accelerating the reaction kinetics. The fuel cell performance strongly depends on the operating conditions; these performances are related to changes in controllable parameters (e.g. pressures, the compositions of the gas, temperatures, current densities, factors using reagents ...) and other less controllable factors talcum impurities, service life.... More, several physical parameters involved in the steering system of the fuel cell. Therefore, it is not simple to establish relationships between the causes that may have an influence on the system and measurable effects, or see if there are interactions between factors. In this context, we will study the influence of flows rates and pressures of the reactant gases (hydrogen and air) on maximum power and performance of the SOFC by using the Design of Experiment methodology, which achieves a better knowledge of behavior of the SOFC system in the face of different factors that are likely to change and that, a minimum testing and with maximum precision.***

Keywords: Solid Oxide Fuel Cell, modeling, Theoretical Model, SOFC, Hydrogen

1. Introduction

Protecting the environment requires reducing emissions of greenhouse gases. In last years, the increase of oil price is the main reasons why researchers and industrialists are pushing to find new solutions for the future. Fuel cells can produce electricity from natural gas, they theoretically reject only water and heat, the ideal configuration in matter of air pollution would be a cell operating on pure hydrogen. The solid oxide fuel cell (SOFC) is intended to the simultaneous production of electricity and heat (cogeneration), it works at the highest temperature (700 °C to 1000 °C) which is required to allow the solid electrolyte (eg. ceramic electrolyte) to have a sufficient proton conductivity, and thus accelerating the reaction kinetics. SOFC technology is a promising technology for industry in the next ten years as it provides powers of several hundred *kW*.

The cell modeling is a very important phase of research because it contributes to a better understanding and representation of the phenomena involved in the batteries. A change in operating conditions can have a beneficial or detrimental impact on the performance of the cell and system performance. The influences of these operating parameters are often difficult to accurately quantify and not obvious to articulate by mathematical relationships. The approach of the design of experiment methodology often has a major interest in the development of complex technology, such as that of the fuel cell. The goal is of model the behavior of processes and/or products to better predict and improve their performance.

2. Scientific Approach

To optimize the organization of experiments and effectively exploit the results obtained, the scientist may have an interest in the use of methods such as Experimental Design (ED). These methods allow to better organize the tests that accompany scientific research or industrial studies [1]. They are applicable to many disciplines and all industries where the main goal is to find the relationship between a variable of interest, Y and variables X_i. We must think of experimental planes if one is interested in a function such as:

$$Y = f(X_i) \quad (1)$$

The primary objective of the experimental desing method is to obtain maximum information with a minimal number of experiments. To do this, we have to follow mathematical rules and adopt a rigorous approach [2]. Generally, an ED consists of highlighting and quantify the existing influence between two types of variables:

- Factors: variables or states that act on the system studied.
- Response: the measured quantity to ascertain the effects of the factors on the system. The response should, of course, be representative of the observed phenomenon.

All experiments must be the subject of careful planning which is reflected in the form of an experimental plane or experimental protocol. The methodological approach of an ED can be divided into different stages [3].

2.1 Description of the element (The SOFC)

The SOFC is potentially more interesting for its high electrical efficiency (50% to 70%) and a less sensitivity to fuel type [4]. Currently, there are four cell technologies in

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matter of geometry: tubular, conventional planar, monolithic planar and Sulzer Hexis. A SOFC cell includes an anode, an electrolyte, a cathode and interconnections.

- Electrolyte: The electrolyte generally used in the SOFC consists of zirconium oxide ZrO_2 and Ytterbium Y_2O_3. The yttria zirconia is very stable in the oxidation and reduction reactions and has an ionic conductivity in the vicinity of 0.02 Scm^{-1} at 800 °C and 0.1Scm^{-1} at 1000 °C. The ionic conductivity is provided by the mobility of oxygen through the anion of yttria zirconia vacancies.
- Anode: The material used in the anode of the SOFC is a cermet (mixture of porous nickel and YSZ), it has good catalytic activity and is inexpensive. The mixture (nickel-YSZ) can operate at low temperatures near 500 °C.
- Cathode: The cathode materials operate in highly oxidizing conditions (air or oxygen + high temperature) which prohibits the use of conventional materials and requires the use of noble materials (semiconductors oxides, conductive metallic oxides). The most used material for the cathode is a lanthanum manganite doped with strontium.
- Interconnection: The interconnections in the SOFC must play the role of current collector and also be completely hermetic. Due to the high operating temperature, the interconnections must have good resistance to oxidation and reduction reactions. Generally a chromium metal is used.

The electrochemical process of a unit cell can be generally described as given in Fig. 1 The anode is supplied with hydrogen or a mixture gas (fuel) and the cathode is supplied with the air (oxidizer).

For cells with high performance operating at high temperature, the fuel mostly used is gaseous hydrogen. In a SOFC cell, water is produced at the anode. The electrochemical reactions are:

At the anode:
$H_2 + O^{2-} \rightarrow H_2O + 2\,e^-$ (2)
At the cathode:
$O_2 + 4\,e^- \rightarrow 2\,O^{2-}$ (3)
The overall cell reaction is:
$H_2 + ½\,O2 \rightarrow H_2O$ (4)

The passage of O^{2-} ions is performed through the electrolyte in a form more or less hydrated. The electrons collected at the anode join the cathode through the load. The process produces water (from hydrogen and oxygen), electricity and heat.

2.2 Defining the response allowing achieving the goal

Response Y (output variable) will be the electric power (P_{el}) issued by cell. For each energy conversion, it is necessary to quantify the input energy of a system and the energy supplied by the latter. In the case of SOFC, the input energy can be considered the chemical energy contained in the hydrogen gas supplying the cell. The product of the cell current (I_{cell}) by the measured terminal voltage (U_{cell}) calculates the electric power (P_{el}) supplied by cell.

$$P_{el} = U_{cell} \times I_{cell} \quad (5)$$

The goal of the study is to determine the effect of the different factors on the electrical power of the SOFC.

2.3 Definition of influential factors

The input parameters of a system (input variables) are called *X* factors, these are possible causes of variation in the response *Y* [5]. In our case, the factors are the pressures and flow rates of the input gases illustrated in Fig.2. We measure the input factors of the cell which are: hydrogen pressure (PH_2), air pressure (P_{air}), hydrogen flow (FH_2) and air flow (F_{air}). When studying the influence of a factor, in general, its variations is limited, according to Yates notation [6], from a low level indicated by (-1) and high level (+1). The choice of range of study is also constrained by combinations of impossible levels.

2.4 Representation of the experimental table

Representation that uses coded units is more general than that which employs the usual physical units, it is the most often adopted and we will use it in the present work. A full factorial plane is a plane for which all possible combinations in the limits of the study have been performed: it is maximum number of trials for a factorial experimental plane. The number of tests N is calculated from the following formula:

$N = 2^k$ where k is the number of factors.

We summarize the 2^4 trials of the full plane in Table 1, if we look at the maximum power delivered by the cell.

2.5 Experimental modeling

Each experimental point gives a value of the response. This response is fitted by a polynomial whose coefficients are the unknowns to be determined. At the end of experiments, this system can be written in a simple manner in matrix notation [2]:

$$Y = aX + e \quad (6)$$

where:
Y: is the responses vector.

X: the matrix coefficient or model matrix which depends on the experimental points selected to execute the plane and model postulated

a: is the coefficients vector.
e: is the differences vector, which contains the lack of fit and the pure error

For 2^4 full design: 16 experiments is needed to determine 16 unknown (constants, main effects and interactions of order 2, order 3 and order 4)

$$Y = a_0 + a_1X_1 + a_2X_2 + a_3X_3 + a_4X_4 + a_{12}X_1X_2 + a_{13}X_1X_3 + a_{14}X_1X_4 + a_{23}X_2X_3 + a_{24}X_2X_4 + a_{34}X_3X_4 + a_{123}X_1X_2X_3 + a_{124}X_1X_2X_4 + a_{134}X_1X_3X_4 + a_{234}X_2X_3X_4 + a_{1234}X_1X_2X_3X_4 + e \quad (7)$$

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here:
a_0 : value of response at the center of the domain = arithmetic mean of the responses
a_1 : Effect of factor 1, a_2: effect of factor 2, a_3: effect of factor 3, a_4: effect of factor 4.
a_{12}, a_{13},…a_{1234} : are the coefficients of the polynomial effect of the interaction between the factors.
In a full or fractional factorial plane with N trials, the responses *Yi* and the coefficients a_i are values of random variables, that each coefficient a_i of the model is calculated by an analytical*:*

$$a_0 = \frac{\left(\sum_{j=1}^{N} +Y_j\right)}{N}, \; a_i = \frac{\left(\sum_{j=1}^{N} \pm Y_j\right)}{N} \text{ and } Y_i = \sum_1^N \pm a_j \quad (8)$$

The formula (8) is applied to determine, for example, the coefficient a_{24} and can check Y_5

$$a_{24} = \frac{(Y_1 - Y_2 + Y_3 - Y_4 - Y_5 + Y_6 - Y_7 + Y_8 + Y_9 - Y_{10} + Y_{11} - Y_{12} - Y_{13} + Y_{14} - Y_{15} + Y_{16})}{16}$$

$$Y_5 = (a_0 - a_1 + a_2 - a_3 - a_4 - a_{12} + a_{13} + a_{14} - a_{23} - a_{24} + a_{34} + a_{123} + a_{124} - a_{134} - a_{234} - a_{1234})$$

The main disadvantages of full factorial desing is excessive tests to be performed when the number of factors becomes important. Fractional desing uses only a fraction of all possible combinations contained in a full factorial plane, it minimizes the number of experiments in this case, retaining only experiments for which:

$$X_1X_2X_3X_4 = +(E_1) \quad (9)$$

thus obtaining the fractional plane illustrated in Table 1 by colored lines, which are representing 8 experiments
We can then write for the 8 experiments: $X_4 = X_1X_2X_3$. As this equality is true for 8 experiments, we can write the following equality column:

$$X_4 = X_1X_2X_3 \quad (10)$$

Recalling that the product of any variable by itself is always equal to + 1, thus:

$$X_1X_4 = X_2X_3,\ X_2X_4 = X_1X_3,\ X_3X_4 = X_1X_2,\ X_1X_2X_4 = X_3,$$
$$X_1X_3X_4 = X_2,\ X_2X_3X_4 = X_1,\ X_1X_2X_3X_4 = 1 \quad (11)$$

The model (7) can thus be written:

$$Y = (a_0 + a_{1234}) + (a_1 + a_{234})X_1 + (a_2 + a_{134})X_2 + (a_3 + a_{124})X_3 + (a_{12} + a_{34})X_1X_2$$
$$+(a_{13} + a_{24})X_1X_3 + (a_{23} + a_{14})X_2X_3 + (a_4 + a_{123})X_1X_2X_3 + e \quad (12)$$

We can use the matrix effects of a 2^3 full factorial plane to determine the coefficients.

3. Conclusion

- We have seen that the operating temperature of the SOFC is 700-1000 °C, so the cell performance is diminished if there is a lowering of the temperature.
- The SOFC cell creates a very clean energy, and develops higher powers than other types of fuel cell.
- The EP method consists of safe tools to drive with maximum efficiency a study involving many parameters, the method relies on rigorous mathematical foundations.
- The theory of experimental plane provides the conditions for which the best accuracy is obtained with minimal testing. We have therefore maximum efficiency with a minimum of experiments and therefore a minimum cost.

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Figure Captions

Fig. 1 Operating principle of a SOFC hydrogen-water fuel cell
Fig. 2 Factors X_i and response Y of process

Table 1: Full Factorial design 2^4 (the whole table), fractional design (colored lines)

N°		The factors X_i				Interactions										Y
	a_0	$X_1=PH_2$	$X_2=Pair$	$X_3=FH_2$	$X_4=Fair$	X_{12}	X_{13}	X_{14}	X_{23}	X_{24}	X_{34}	X_{123}	X_{124}	X_{134}	X_{234}	X_{1234}	$Y=P_{el}$
1	+1	-1	-1	-1	-1	+	+	+	+	+	+	-	-	-	-	+	Y_1
2	+1	-1	-1	-1	+1	+	+	-	+	-	-	+	+	+	+	-	Y_2
3	+1	-1	-1	+1	-1	+	-	+	-	+	-	+	-	+	+	-	Y_3
4	+1	-1	-1	+1	+1	+	-	-	-	-	+	+	+	-	+	+	Y_4
5	+1	-1	+1	-1	-1	-	+	+	-	-	+	+	+	-	-	-	Y_5
6	+1	-1	+1	-1	+1	-	+	-	-	+	-	+	-	+	+	+	Y_6
7	+1	-1	+1	+1	-1	-	-	+	+	-	-	-	+	+	-	+	Y_7
8	+1	-1	+1	+1	+1	-	-	-	+	+	+	-	-	-	-	-	Y_8
9	+1	+1	-1	-1	-1	-	-	-	+	+	+	+	+	+	+	-	Y_9
10	+1	+1	-1	-1	+1	-	-	+	+	-	-	+	-	-	-	+	Y_{10}
11	+1	+1	-1	+1	-1	-	+	-	-	+	-	-	+	-	+	+	Y_{11}
12	+1	+1	-1	+1	+1	-	+	+	-	-	+	-	-	+	-	-	Y_{12}
13	+1	+1	+1	-1	-1	+	-	-	-	-	+	-	-	+	+	+	Y_{13}
14	+1	+1	+1	-1	+1	+	-	+	-	+	-	-	+	-	-	-	Y_{14}
15	+1	+1	+1	+1	-1	+	+	-	+	-	-	+	-	-	-	-	Y_{15}
16	+1	+1	+1	+1	+1	+	+	+	+	+	+	+	+	+	+	+	Y_{16}
	a_0	a_1	a_2	a_3	a_4	a_{12}	a_{13}	a_{14}	a_{23}	a_{24}	a_{34}	a_{123}	a_{124}	a_{134}	a_{234}	a_{1234}	

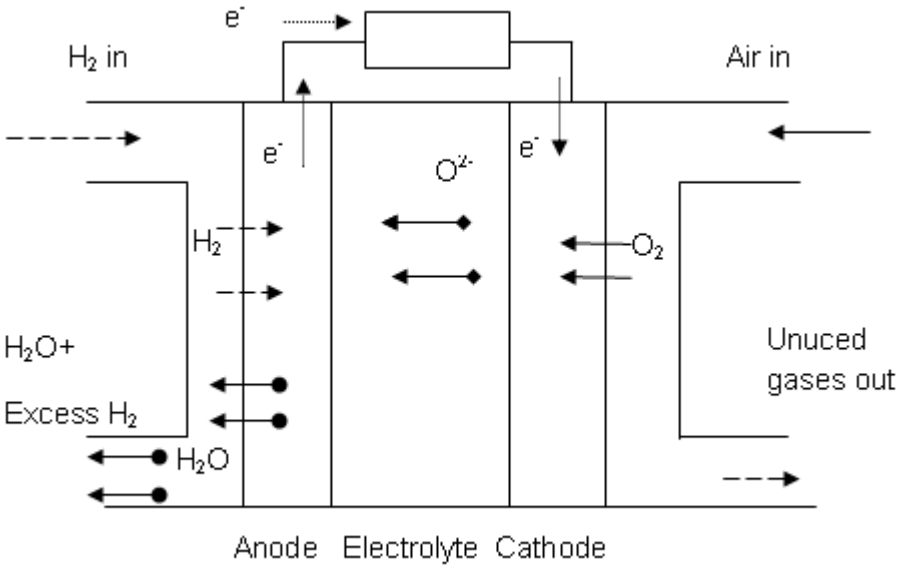

Figure 1: Operating principle of a SOFC hydrogen-water fuel cell

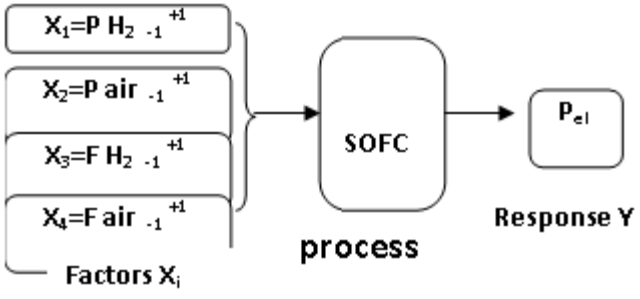

Figure 2: Factors X_i and response Y of process

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