

EXPERIMENTAL EVALUATION OF REFERENCE VALUES FOR PEM FUEL CELLS

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In this work, we carry out detailed investigations on the performance of a home-built 92cm² PEMFC under specific operational conditions, such as humidity, temperature, air back pressure, stoichiometry, and gas diffusion layer deformation. We show that the optimal working point (i.e., 0.6V/38A) is consistent with high levels of humidification (i.e., 95%) and gas diffusion layer deformation (i.e., ~45%), at temperatures around 55 °C. The obtained results are in good agreement with mathematical modeling by using finite element calculation methods.

Keywords: fuel cell, proton exchange membrane, polarization curve

1. Introduction

Among various known green technologies, the proton exchange membrane (PEM) fuel cell has been one of the most challenging and problematic issue. The concept of PEM fuel cells replacing batteries is often debated in terms of energy density, technological feasibility, safety and costs. It turns out that fuel cells are best suited to applications adaptable to customer demand that involve high power density, stability and prolonged life cycle. Fuel cells in the form of batteries are, however, attractive since they offer a great deal of versatility and degree of freedom [1]. These features become increasingly relevant with, for instance, the emergent technologies that are interlinked via independent or self-sustained energy sources capable to deliver high power densities instantaneously.

In contrast to batteries, since the energy source and the energy converter are separated, a fuel cell system adds complexity and associated safety, whereas reliability issues will need to be carefully assessed for each application in part. The prospective commercial market for high energy density power sources is attractive enough to support significant development and to accelerate the introduction of small and medium fuel cells since battery technology is unlikely to be able to meet the growing energy demands of a mobile workforce [2].

One key issue to integrating fuel cells in power application is the use of the so called *proton exchange membrane fuel cells* (PEMFC). Some advantages of

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PEMFC over the other fuel cells is given by their low operating temperature, sustained operation at a high current densities, low weight, compactness, the potential for low cost and volume, long stack life, fast start-ups and suitability for discontinuous operation [2-5]. These features make PEMFCs the most promising and attractive candidate for a wide variety of power applications ranging from portable and wearable [6], and transportation to large-scale stationary power systems for buildings and distributed generation.

In this work, we look in detail at the functional parameters of a home-built PEM fuel cell. First, we describe the fundamental aspects related to the functional principle on which a fuel cell is based. Together with that, the components involved in the fabrication of our fuel cell are presented and their specific role is discussed. Next we show how the operating conditions influence the overall response of our fuel cell. Explicitly, we point out the influence of *i*) oxidant and fuel humidity, *ii*) cell temperature, *iii*) internal level of GDL deformation, *iv*) the overall pressure drop along the cell, and *v*) the influence of oxidant-to-fuel ratio (i.e., stoichiometry). Each of these aspects are discussed separately, whereas their merged contribution to the overall power output of our fuel cell is emphasized by comparing the experimental results to theoretical polarization curves obtained from simulations by using dedicated finite element computational software (i.e., Matlab and Fluent).

2. Polymer Electrolyte Membrane Fuel Cells

A typical polymer electrolyte membrane fuel cell is based on the membrane electrode assembly (MEA), which is the living core of a fuel cell. It consists of a polymer (e.g., Nafion) with proton selective conduction properties from one side (i.e., that is anode) to the other (cathode), upon application of a potential difference. In order to achieve that, however, a thin porous electrode of carbon (e.g., carbon black) is deposited on the proton conducting polymer and loaded with no more than 0.4 g/cm^2 Pt catalyst whose role is to promote hydrogen dissociation and absorption from gaseous atmosphere. In order to provide both the oxidant and fuel gas together with the water drain management as product of reaction, especially engraved carbon plates (i.e., bipolar plates) are used in conjunction with a highly porous gas diffusion layer (GDL), whose role is also to ensure excellent electrical contacts. The system is mounting between copper plates to facilitate electrical current collection at the end of the cell that is robustly enclosed between two stainless-steel plates equipped with inlets and outlets for gas and water flow management. A more detailed description is given in Fig. 1 a).

The choice for a specific channel flow geometry comes in accordance to scaling PEMFCs, in other words it depends on whether high or low pressure drops across MEA influences dramatically water removal from flooded channels or not.

In order to circumvent that, we have suggested that the parallel serpentine flow geometry (i.e., in Fig.1, b) is best suited to our demands since oxidant and fuel gasses are prepared externally in high capacity moisturizers at temperatures above the working temperature of our PEMFC so as water removal from drained channels, either due to dew point gradients across the cell or as a direct result of reaction, is effective.

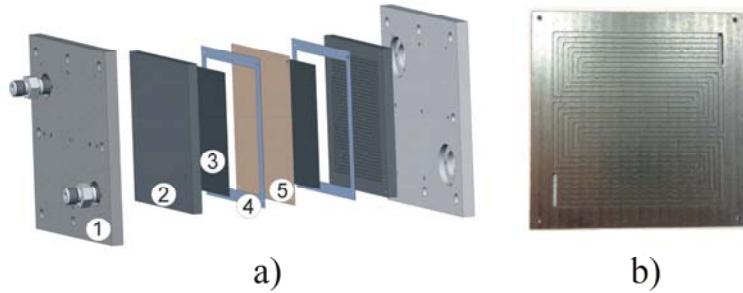


Fig 1: Schematic representation of PEMFC hierarchy. In a), membrane electrode assembly, MEA (5) that separates anode from cathode is sealed by fast prototyping gaskets (4) that also inner-host a highly porous gas diffusion layer, GDL (3) whose role is to facilitate the water management and to insure electrical contact with the bipolar plates, BP (2). Although not shown, copper plates placed between BP and the end stainless-steel plates (1) are used to collect the electrical currents. In b), an illustration of the parallel serpentine flow and water drain channels engraved on the bipolar plates is shown

In this work, we use commercially available MEA (i.e., with active area of 92cm^2), comprising of a 0.018 mm thick proton conducting polymer (Nafion) capped on each side with ~ 0.010 mm thick electrodes of carbon black, loaded unequally with Pt catalyst. For gaskets, we make use of Polyolefin elastomers with personalized topography to compensate for component tolerances by up to 55% in height deformation. This specific design is especially suited for fast PEMFC prototyping, ensuring handling with ease and enhanced durability upon many closing/opening cycles for a wide range of applied stress and operating temperatures. As gas diffusion layer (GDL), two 92cm^2 and 0.30mm thick carbon paper sheets from GDL34BC Sigracet with areal weight of 140 g/m^2 , 75% porosity and through-plane electrical resistivity $<14\text{m}\Omega\text{cm}^2$. The bipolar plates are made out of 3.00 mm thick carbon plates (i.e., from Graftech, FFP200) with low density (i.e., 1.50 g/cm^3), high tensile and flexural (i.e., $> 30\text{ MPa}$ and $>50\text{MPa}$, respectively), low *in-* and *out-of-plane* electrical resistivities (i.e., 7 and $300\text{ }\mu\Omega\text{cm}$, respectively), and excellent thermal conductivity (i.e., $>275\text{ W/mK}$). The engraved parallel serpentine flow delineates 7 channels with width of 1.4 mm and height of 0.5mm, separated by 1.4 mm wide lands, as shown in Fig 1. b). The

current collector plates are made of 2.5 mm thick gold electroplated copper plates. The cell is closed between 12 mm thick W.1.4306 Stainless Steel end plates equipped with gas flow and water drain Swagelok fittings.

3. Test stand for PEM fuel cells

To fully characterize our PEMFC, we first close the cell at a GDL deformation of 7%. The fuel cell is then mounted on our home-built testing stand, whose block diagram is illustrated in Fig. 2. Dry fuel and air are fed via PVC pipes, which are equipped with pressure gauges and flow controllers (i.e., either in mass or volume) in order to quantify fuel cell consumption and to regulate the stoichiometry in operational mode. Two identical moisturizing bubble humidifiers are used to humidify the gases. This can be done up to 45% at 25 °C and 95% at 71 °C. The overall pressure drop over both the anode and cathode is set by using back-up pressure regulators, up to 1.3bar absolute pressure. Two identical heating elements are mounted on both electrodes to set and control cell temperature, up to 71 °C. When temperature, stoichiometry, back-pressure and flow rates are set, an I-V curve is measured between the open circuit voltage value down to 0.3 V. That is carried out by using a PLA800 60-300 electrical load bridge (i.e., from AMREL American Reliance Inc.) that is able to drain 300A up to 60V appliance. This characterization sequence is repeated at 15%, 25%, 35%, 45% and 55% level of deformation with respect to GDL.

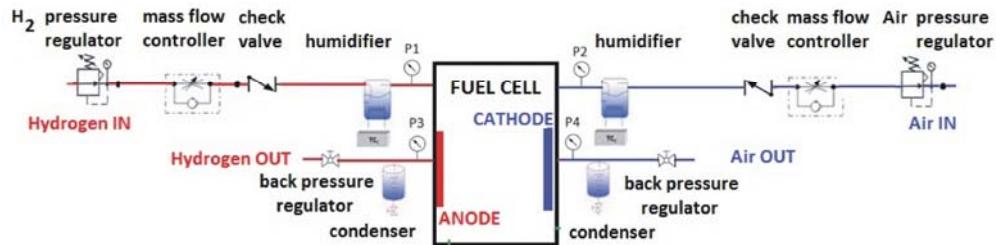


Fig 2: Schematic representation of the block diagram used to characterize PEMFC. The corresponding pneumatic lines for the anode and cathode sides are shown, respectively

4. Operation principle of PEM fuel cells

Humidified H₂ gas is supplied under pressure into the anode gas channel and diffuses through GDL until it reaches the catalyst layer (i.e., in Fig. 3 a). In the presence of Pt catalyst, hydrogen molecules dissociate according to the following reaction:



The resulted protons (i.e., H^+) are transferred through the proton conducting membrane to the cathode; whereas the electrons are pass the external circuit to the cathode. Similarly, humidified oxygen (or air) is supplied into the cathode and diffuses through the porous GDL to reduce at the Pt catalyst layer into two oxygen atoms. They combine with protons, driven through the proton conducting membrane, and with electrons traveling through the external circuit and arriving at the cathode to produce water:



In addition to water, heat is also produced in this reaction. The overall electrochemical reaction occurring in the PEM fuel cell is given by the following reaction:



A PEM fuel cell operating at 25 °C and 1 atm, the potential difference across the cell would be about 1.23 V. That is known as reversible cell potential. Due to various losses, also known as *polarizations* or *overpotentials*, the actual cell voltage is always lower than the theoretical value. The losses result from three main sources, *i*) *activation overpotentials* due to the kinetics of charge transfer reactions in anode and cathode catalyst layers, *ii*) *ohmic overpotentials* due to the resistances of cell components, and *iii*) *concentration overpotentials* due to the limited rate of mass transfer. These features are illustrated in Fig. 3 b).

For the polarization curve in Fig. 3 b), Kim *et al.* [7] developed an empirical relation which takes into account an exponential term for the mass-transport overpotential with an adjustable parametric coefficient. This accurately describes the performance curves, including the mass-transport limited region at high current densities. The cell voltage is calculated using the following relation:

$$E_{cell} = E^{rev} - \eta_{act} - \eta_{ohm} - \eta_{conc} \quad (4)$$

$$= E_0 - b \log \frac{J}{J_0} - RJ - m \exp(nJ) \quad (5)$$

where last three terms in equation (4) account for the activation, ohmic, and concentration overpotentials, respectively. J is the cell current density, and b , J_0 , R , m and n are empirical parameters. E^{rev} is the *open circuit potential value*. By using equation (5), all PEMFC polarization curves can be accurately described. The main differences between, say, two polarization curves measured in different conditions arise mainly from ohmic losses (i.e., in eq. (5), the $-RJ$ term). This is, however, due to the fact that the functional parameters of PEM fuel cells are

always set for this regime, which is the optimal work regime where the relation between the fuel-to-oxidant consumption is proportional to the output electrical power.

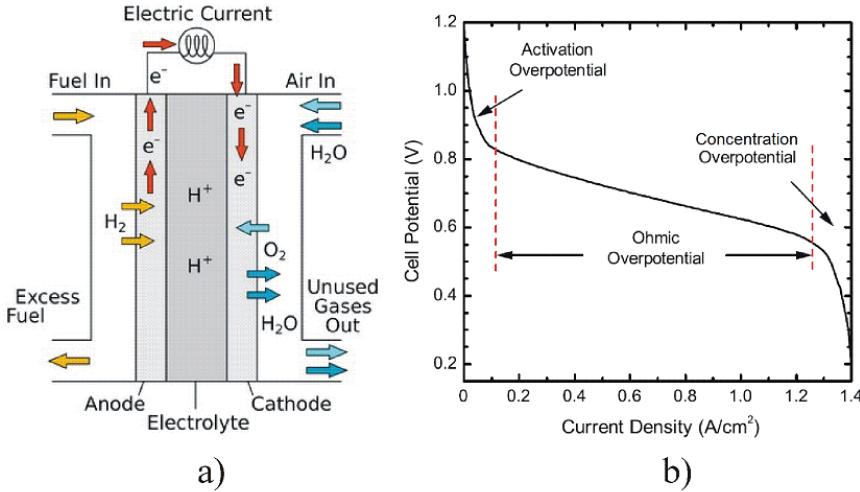


Fig 3: Schematic representation of the electrochemical processes in PEM fuel cells. In a), the anode and cathode reactions (i.e., according to equations (1) and (2), respectively) mediated by proton transfer through MEA take part to produce electrical charge that is used in an external circuit (source: Wikipedia). In b), the relation between the working potential of a PEM fuel cell is shown as a function of the drained electrical current. The specific contributions to that are indicated.

For the ohmic regime (e.g., in Fig. 3b), the slope value for the I-V curve is given by the contact resistance of the inner components of the fuel cell, accounting for all interfaces and the proton conducting membrane as well. Part of the overall ohmic losses in PEMFCs arises from physical contact among the constitutive materials. This may be responsible for up to 45% of cell ohmic losses, whereas nearly $\sim 55\%$ is due to membrane itself. One should, however, keep in mind that the membrane conductivity is relatively poor (e.g., ~ 0.1 S/cm for a 95% RH membrane at 80°C) compared to the conductivity of the other constitutive parts of the fuel cell (e.g., the carbon plates have a much higher conductivity of ~ 5000 S/cm).

5. Results and Discussion

We show now the performance exhibited by our 92 cm^2 PEMFC under various imposed operation conditions, such as: humidity, temperature, air back-pressure, stoichiometry. Each working parameter has a certain influence on the electrical current drained out from the fuel cell and, to some extent, their overall contribution to the fuel cell performance can be emphasized. For instance, the role

of humidity is to ensure an increased proton conductivity of the polymer membrane, whereas the cell temperature and gas pressure and stoichiometry have over imposed contributions given by Nernst equation that takes into account both the fuel and oxidant species. Additionally, the level of GDL deformation, which is related to the clamping force on the fuel cell, has its own influence on the fuel cell performance since higher deformations result in better electrical contacts between the constitutive parts of the cell. Next, we describe these contributions in part.

5.1. Influence of humidity

The fact that the supplied oxidant and fuel gasses need to be humidified prior to feeding the fuel cell comes in accordance with the enhanced proton conductivity of the electrolytic membrane with increasing water content and temperature (i.e., hopping term). This is given by the parametric relation:

$$\sigma(\lambda, T) = (0.5139 \cdot \lambda - 0.3260) \cdot e^{1268(\frac{1}{303} - \frac{1}{T})} \quad (6)$$

where T is temperature (in K) and λ a parameter whose value is between 1 and 14, when the membrane humidity is varied between 10% and 100%, respectively. More details on that may be found in Ref. [4].

The influence of humidity on the fuel cell performance is shown in Fig. 4a). The cell is closed at 7% GDL deformation, the temperature is set to 55°C and the moisturizing vessels heated such as the humidity level of hydrogen and air is dry (i.e., 40% RH) and wet (i.e., 95% RH). The first feature exhibited by the I-V characteristics is the relatively low current densities obtained. This is due to the low compression strength on the fuel cells which results in relatively high ohmic losses. However, it is clear that the fuel cell performs better when fed with wet reactants. This can be understood in terms of an enhanced proton conductivity of the polymer membrane, as described by eq. (6). In order to emphasize that, we have carried out similar experiments at 23, 41, 55 and 71°C, whereas the humidity levels are kept the same. The corresponding ohmic regimes are fit to linear expressions to obtain the reduced specific conductance values (i.e., the inverted value of resistance from the term $-RJ$ in eq. (5), with J expresses in A/cm²). The results are shown on a logarithm scale in the inset of Fig. 4a), as a function of the inverted temperature (i.e., Arrhenius plot). Note that for the dry (i.e., 40% RH) conditions used in this work, the fuel cell conductance drops at high temperatures (i.e., at 71°C) as a result of membrane dry out. As we shall see, this is typical for the membrane electrode assembly used in this work.

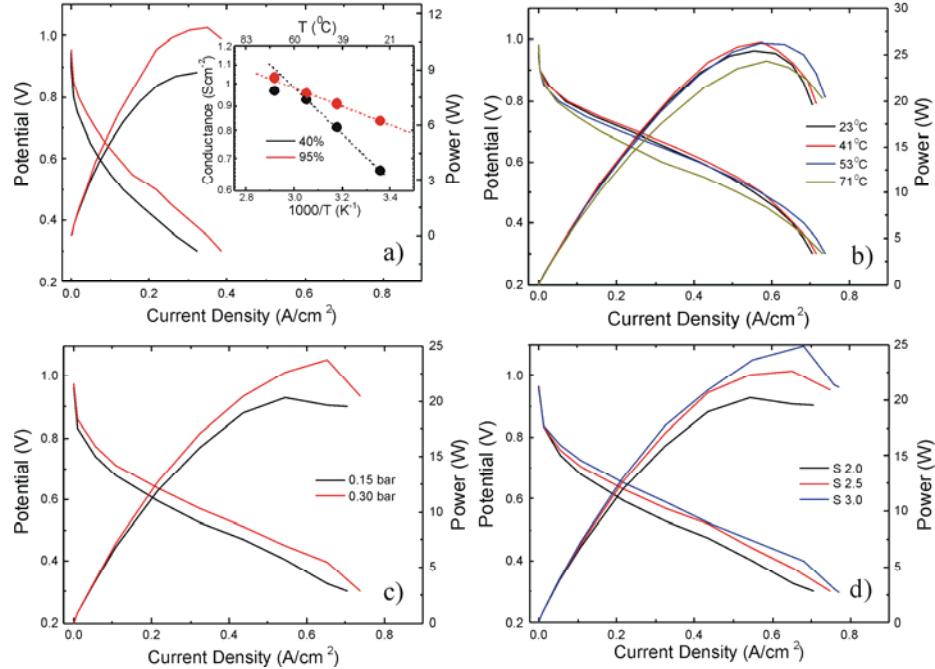


Fig 4: Experimental evaluation of the I-V characteristics and power delivered by our 92cm² PEMFC under various working conditions of humidity, temperature, air pressure and stoichiometry. In a), the influence of humidity (i.e., for RH values of 40% and 95%) on the performance of our PEMFC closed at 7% GDL deformation is shown. The inset represents the Arrhenius plot of the inverted reduced resistance (i.e., or conductance) determined from fits to the corresponding data by using eq. (5). In b), c) and d), the influence of temperature, air pressure and stoichiometry PEMFC performance closed at 45% GDL deformation are represented, respectively.

5.2. Influence of temperature

The influence of temperature on the fuel cell performance is shown in Fig. 4b). As a matter of fact, a glimpse on that is given already in the inset of Fig. 4a). However, the difference in Fig. 4b) is the increased GDL deformation level (i.e., 45%). This is depicted from the increased current densities and maximum power obtained at all temperatures, whereas the gas humidity is set to 95%. Up to 55°C, the fuel cell performance increases gradually with increasing temperature in a manner similar to that shown in the inset of Fig. 4a). This is followed by a sudden decrease of cell performance at higher temperatures (i.e., at 71°C). Most probably, this is due to cell dry out.

5.3. Influence of pressure

In order to understand the influence of gas pressure on fuel cell performance, one should, however, keep in mind that hydrogen and air have very

different diffusivity values when propagating through porous media, such as GDL [4]. Hydrogen is able to fill any space fast whereas oxygen diffuses slower due to the increased molecular radius and mass. Therefore, one would expect that the PEMFC performance is influenced by air pressure management rather than the hydrogen one. To that, water drainage as a reaction product at the cathode side plays an important role since its presence lowers the active area reached by oxygen and it mediates the temperature gradients along the cathode flow channels. This is, nevertheless, a very delicate thermodynamic balance within the grain boundaries of a multi-phase system that consists of wet gasses, liquid water and steam, according to the Nernst equation for the open circuit potential E^{rev} from eq. (4); i.e.,:

$$E^{rev} = E^0 + \frac{RT}{2F} \cdot \ln\left(\frac{pH_2 \cdot \sqrt{pO_2}}{pH_2O}\right) \quad (7)$$

where pH_2 , pO_2 and pH_2O are the partial pressures of hydrogen, oxygen (i.e., in air) and steam water, respectively; R is the ideal gas constant (i.e., 8.314 J/molK), F the Faraday constant (i.e., 9648.53 C/mol) and T temperature (in K). From eq. (7), it becomes clear that a fuel cell is typically operated at elevated pressures to ensure proper flow of reactants to the electrodes. This is also to increase the reaction kinetics at the cathode side by increasing the open circuit potential value.

The influence of back pressure (i.e., on the cathode side) is shown in Fig. 4c). The experiments are carried out at 55°C on a PEMFC closed at 45% GDL deformation and humidified at 95% RH. The data indicate clearly that the higher the pressure along the cathode flow channel, the better the fuel cell performance in both, the current densities values and the maximum output power.

5.4. Influence of stoichiometry

The influence of stoichiometry on the fuel cell performance is shown in Fig. 4d). Stoichiometry refers at the molar ratio of oxygen with respect to hydrogen, a stoichiometry value of 2 meaning that 2 protons combine with 1 of oxygen to result one mole of water. Typically, due to the fact that oxygen diffuses slower through the porous GDL than the hydrogen gas, PEMFC are fed with oxygen (air) in excess. Higher stoichiometry values lead to an increased fuel cell performance as more oxidant is able to make it through the cathode flow channels to the membrane electrode assembly.

5.5. Influence of GDL deformation

The overall performance exhibited by PEMFC as a function of humidity, temperature, back pressure and stoichiometry may be further improved by reducing the ohmic losses that are responsible for more than 50% of the output current density and power values. This can be achieved by optimizing the GDL

deformation level, compressing the GDL structure from initial thickness value down to optimal one. The measurements were done with special calibers inserted between fuel cell end plates. This can be easily understood by comparing, for instance, the current density and the power output obtained on our PEMFC closed at 7% and 45% GDL deformation level, as shown in Fig. 4a) and b), respectively. Since these features exhibit dramatic changes, we have conducted a detailed study on the influence of GDL compression on the fuel cell performance when operated at 55 °C under 95% humidification and nearly 2.0 stoichiometry value. The results shown in Fig. 5 indicate clearly that the best performance of our fuel cell is obtained for a GDL deformation level of 45%. Nevertheless, higher deformations of GDL result in reduced pore size which influences in turn gas and water management along the diffusion layer. In Fig. 5, this corresponds to a lower power output for 55% GDL compression.

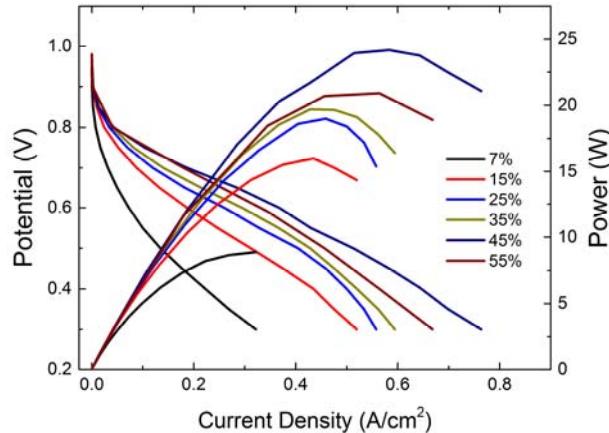


Fig 5: Performance of a 92cm² PEMFC closed at various levels of GDL deformation. The fuel cell is operated at 55°C, under 95 % humidity and nearly 2.0 stoichiometry value. The best performance is obtained at 45% GDL deformation level.

6. Mathematical Modeling

Mathematical modelling of PEMFC, based on an accurate description of the associated processes, is an indispensable tool for exploring various architectures for fuel cells and their components and also for parameter optimization. In this work, the modelling approach is based on a complete set of mathematical equations accounting for fluid flow, multi-component species transport and electrochemistry.

The performance of our PEMFC is studied using a single-phase, steady state, three-dimensional electrochemical model. Physical and electrochemical

phenomena that take place inside a fuel cell are represented by mathematical models. The data analysis is based on solutions for the mass, momentum, species and charge transport equations. The physical laws involved in this study account for *i*) fluid flow in porous media (i.e., Darcy equations for GDL and catalyst layers), *ii*) multiphase diffusion (i.e., Stefan-Maxwell equation), *iii*) faradaic charge transfer (i.e., Butler-Volmer equation) and *iv*) ohmic losses (i.e., Ohm law). However, in order to simplify the model, some assumptions are made. Among these, the most used ones are: ideal gas mixtures with ideal gas properties, incompressible laminar flow for the entire cell, isotropic and homogenous membrane and electrode structures, negligible ohmic potential drop in solid components, and application of volume-averaged conservation equations.

Our model takes into account electrochemical kinetics and multi-component species transport for a full PEMFC architecture. This includes all PEMFC components; i.e., flow channels, gas diffusion electrodes, catalyst layers and the polymeric membrane. The transport and electrochemical equations are solved for a single domain. The interfacial boundary conditions between cell components are disregarded.

The PEMFC model is implemented by using a computational fluid dynamics code – ANSYS MULTIPHYSICS. For that, we use an unstructured grid with 1.5 million cells developed with Gambit. A control volume method is used to solve the main equations in ANSYS Fluent with Fuel Cell Module. The solution is based on the SIMPLE algorithm, with a convergence limit of 10^{-5} . All simulations are carried out on computational unit equipped with Intel Core Duo CPU with 3.17GHz and 8GB RAM. The convergence is reached after 800 iterations (i.e., work time of ~30 hours).

In Fig. 6 we show the contour maps for the current density, water content of the membrane, temperature gradients and air pressure along the cathode channels of our PEMFC, operated at 0.6V, 95% RH, 55°C and air back pressure of 0.15bar. The results should be regarded as stationary solutions with respect to the imposed working conditions. For instance, the current density map in a) shows that higher currents are drained at the fuel cell inlet than at the outlet. This is consistent with the increased amount of hydrogen and air fed at the inlet side. However, the overall current density obtained on the entire active area of the fuel cell is $\sim 0.40\text{A/cm}^2$. This value is in good agreement with the experimental imposed conditions.

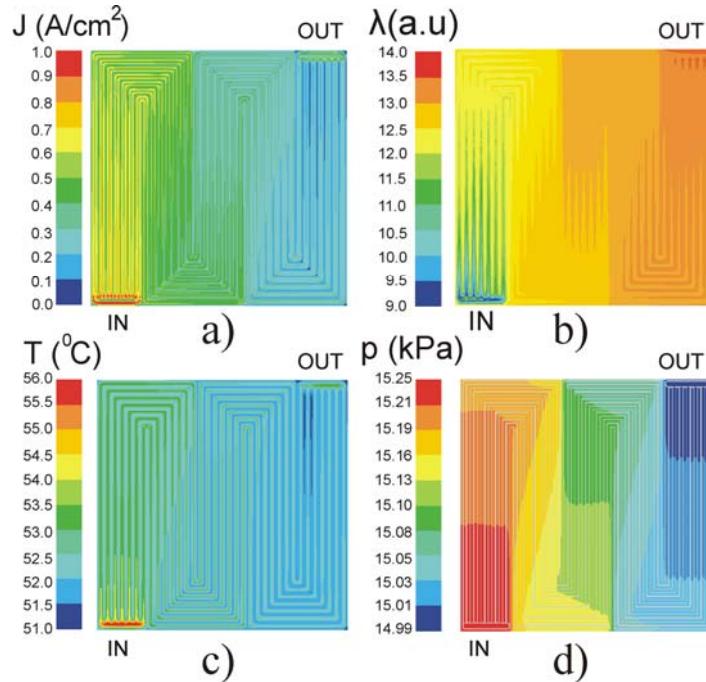


Fig 6: FLUENT resolved contour maps for the current density (in a), water content of the polymer membrane (in b), temperature gradients (in c) and air pressure drop (in d) across the PEMFC drain channels (cathode side). The data correspond to the nominal working point of the PEMFC (i.e., $0.6V / 0.4A/cm^2$), 95% RH at $55^{\circ}C$, with a pressure over the cathode side of 0.15bar air.

In Fig. 6b), we show that the polymer membrane is highly humidified; i.e., the average values for λ in eq. (6) is 12.5. This corresponds to a RH value higher than 85%. Nevertheless, the temperature across the cell shown in Fig. 6c) turns out to be constant (within $\pm 2^{\circ}C$), around the average value of $53^{\circ}C$. Finally, the air pressure drop along the cathode channels is nearly constant (i.e., in Fig. 6d, $p \sim 0.15$ bar). This is consistent with the fact that the air pressure is back regulated with respect to the upstream air flow fed to the fuel cell.

The COMSOL/Fluent simulations return the stationary solutions for the functional parameters at given values for the current density and working potential, respectively. In our case, we carry out the convergence sequence described above at few more working potentials. The results for the resulting I-V characteristic are shown in Fig. 7 together with the experimental data.

The data are in good agreement. However, we fit the experimental data by using the empirical equation (5).

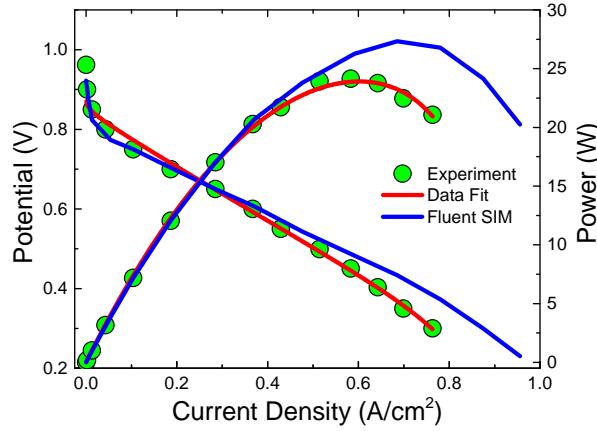


Fig 7: Typical I-V and power characteristics for our PEMFC (i.e., 45% GDL deformation, 95%RH at 55^0C and 0.15bar air). The experimental data are fit by using empirical equation (5), according to REF[8]. The blue curve represents the COMSOLE/FLUENT simulations data point obtained by using the experimental operation conditions (i.e., open circuit voltage, ohmic loss, temperature, humidity and air pressure drop)

For that, we fix the open circuit voltage (OCV) value to the experimental one (i.e., 0.953V), b to 0.014V (i.e., corresponding to $RT/2F$, with $T=328\text{K}$, $R=8.314 \text{ J/molK}$ and $F=9648.53 \text{ C/mol}$) to obtain the exchange current density $J_0=1.0\times10^{-9} \text{ A/cm}^2$ in the Tafel term, and $R = 0.655 \Omega\text{cm}^2$ for the ohmic losses. These values are in good agreement with electrochemical impedance evaluation of the contact resistance value of $7.1 \text{ m}\Omega$.

7. Conclusions

In this work we carry out detailed investigations on a 92 cm^2 PEMFC performance. For that, we point out the influence of the humidity, temperature, air back pressure and stoichiometry on the overall features exhibited by PEMFCs. We find out that all these functional parameters have to be carefully tuned to optimize PEMFC performance, in a way that ensures a high humidification of the polymer membrane, low temperature gradients along cell, constant cathode flow of air to drain out the water excess at fixed stoichiometry values. However, the best performance for our home designed PEMFC is obtained for 95% RH, $T = 55^0\text{C}$, 0.30bar air back pressure and 3.0 stoichiometry. Nevertheless, all these parameters are influenced drastically by the level of deformation of GDL (i.e., 45%). This has double influence on the PEMFC performance: *a positive influence* since high GDL deformations ensure good electrical contact between the constitutive parts of the cell as a result of the increased clamping force, and *a negative one*, due to a decreased GDL porosity which adds resistance to removal

of water product. These features are, however, in good agreement with mathematical modeling of PEMFC which returns consistent values for the operational parameters used in this work.

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