DESIGN CRITERIA OPTIMIZING THE USE OF FUEL CELL SOURCE IN ELECTRIC POWER SYSTEM

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Abstract: Fuel Cell (FC) sources are able to provide electric energy efficiently while respecting environmental constraints. FC systems are basically constituted by a FC stack and a converter, able to adapt the operating conditions to the requirements of the electrical loads. Due to the present FC cost, which reduces the FC application fundamentally to prototype systems, design criteria for the applications of FC sources in electric power systems must be defined in terms of cost minimization. The paper, by analyzing the operational conditions, discusses on the FC behavior, examining the availabilities of electrical energy and power by means of different FC stack configurations. The final aim of the paper is to determine design criteria suitable to suit the FC stack configurations both according to the electrical load requirements and minimizing the overall system cost. *Copyright* © 2005 IFAC

Keywords: Design Criteria; Fuel Cell; Modeling; Optimal configuration; PEM; Static operation.

1. INTRODUCTION

Fuel Cell (FC) sources are able to provide electric energy efficiently while respecting environmental constraints. These characteristics and the possibility of refueling the cell with different fuels are making for an interest in electric power applications sources. In the sector of electric power generation, deregulamentation, power quality and environment issues are increasing the research into using fuel efficiently; in the field of Electric Vehicles design, the research of on-board electrical sources characterized by high energy density seems to favor the analysis and application of FC sources with respect to electrochemical accumulator sources. Because of this, several manufacturers are currently intensively developing FC systems, basically constituted by a FC electrical source and a converter, able to adapt the FC operating conditions to the requirements of the electrical system, Fig. 1 (Cheng, et al., 2001; Santi, et al., 2002).

FC technology is not new, but only as some of the fundamental difficulties have been overcome recently, have FC sources become more feasible and

are gradually being commercialized. Today the limit for the commercialization of FC is the cost, which reduces the application fundamentally to prototype systems. As a consequence, design criteria for the applications of FC sources in electric power systems do not leave out of consideration of optimizing the FC design. In particular, by analyzing FC behaviour, expressed in terms of electrical energy and power, it seems possible to design source configurations able to minimize the system cost (mainly related to the FC active area) however according to the loads requirements. Due to the low cell voltage, FC



Fig. 1. Electric power system FC source-based

sources designed for electrical power applications are assembled in series and/or parallel configurations. Hence, by means of evaluating how the source performances vary in different configurations, it is finally possible to design with minimal cost FC systems, which is suitable to satisfy the electrical load requirements.

In the paper the operation characteristics of a polymeric electrolyte membrane (PEM) FC are investigated. In order to characterize the FC electrical behaviour, the steady-state electrochemical model is defined and the model is analyzed by means of a sensitivity analysis; in particular, according to the defined model, the incidence of the configuration design on system performances are examined. Finally, a series of considerations focusing on FC system design criteria for the applications in electric power system are reported.

2. PEM PHYSICAL OPERATION

The proton exchange membrane fuel cell, PEMFC, takes its name from the special plastic membrane used as electrolyte. It is a perfluorinated sulfonic membrane, characterized by an elevated proton conductibility. The FC combines in a very compact unit the electrodes (anode and cathode) and the electrolyte. This structure, known as Membrane Electrode Assembly (MEA), is no thicker than a few hundred microns. It is the PEM core and, supplied with fuel and air, generates electric power at about 1 V cell voltage, with a power density of up to about 1 W/cm², Fig. 2 (Ronchetti and Iacobazzi, 2002).



Fig. 2. PEMFC structure

The solid polymer electrolyte forms a thin electronic insulator and gas barrier between the two electrodes, allowing fast proton transport and high current densities, as required in electric power applications. Moreover, the solid electrolyte does not diffuse or evaporate, making intermittent operation and rapid load change possible.

The proton transfer in the solid polymer electrolyte follows two principal mechanisms (Gregor, 2002; Mann, *et al.*, 2000). The most trivial case requires the translational dynamics of bigger species and the proton diffuses through the medium together with a "vehicle" (for example as H_3O^+ in the case of H_2O); in the other mechanism, the vehicles show pronounced local dynamics and the protons are transferred from one vehicle to the other by hydrogen bonds. Simultaneous reorganization of the

proton environment, consisting of reorientation of individual species or even more extended ensembles, leads in the formation of an uninterrupted path for proton migration. The mechanism is known as "proton hopping".

The Nafion membrane thickness varies from tens to hundreds of μ m (Laughton, 2002); its proton conductivity is strongly dependent on the level of hydration. When proton exchange membranes are subjected to temperatures above 100 °C at atmospheric pressure, their conductivity decreases significantly due to dehydration: so the operating temperature is limited under 100 °C. Electrocatalysts are required to obtain good performances; the electrocatalysts increase chemical reaction speed and reagents can react with a lower activation energy.

Modern FC electrodes are Gas Diffusion Electrodes (GDEs). They consist of a gas porous layer of high surface area catalyst on a carbon paper or carbon cloth support and a gas porous, electrically conducting gas diffusion layer, or electrode substrate. Two similar electrodes, anode and cathode, are in intimate contact with the polymer electrolyte membrane. As is evident in Fig. 3, a substrate material (normally carbon fiber papers) is used. This substrate, needed as a spacer to allow gas access even to catalyst areas underneath the supporting ribs of the gas distribution structure or flow field, constitutes a unit with minimal ohmic losses. These regions are also known as the "landing area".



Proton Exchange M. Catalyst Layer Electrode Substrate Open Gas Channel Flow Field Plate

Fig. 3. Electrode structure

MEA is located between two plates; these are usually made of graphite, into which are flow channels used to provide the reagents to the anode and cathode. The plate has high electronic and good thermal conductivity and is stable in the chemical environment inside the FC. The thickness is usually of several mm, allowing the engraving of the flow channels. In order to connect the cells in series and to assemble stack, constituted by a sequence of alternative plates, the plate, normally called "bipolar", presents flow fields on the right and the left sides, resulting as cathode for one side and anode for the other. This approach allows the greatest possible flexibility for designing and optimizing the flow field.

Stack components include also cooling elements, current collector, and humidifiers. The cooling maintains the required operating conditions, in order to favour water removal, whereas the humidifier maintains the membrane hydration, for proton conductivity.

The electrochemical reactions, which mainly occur in a PEM, are:

$$H_2 \rightarrow 2H^+ + 2e^- \quad E = 0 \tag{1}$$

$$1/2O_2 + 2H^+ + 2e^- \rightarrow H_2O \quad E = 1.23V$$
 (2)

These electrochemical reactions take place at the anode and the cathode catalyst layer, respectively. At the anode the gas diffusion layer allows hydrogen to reach the reactive zone within the electrode; at the cathode, water is formed according to (2). The membrane acts as a proton conductor; the protons of sulphuric groups (SO₃H) are able to migrate along the molecule of polymer and to transfer the ions H⁺ between the two sides of the membrane.

Stack dimensions depend on the number of cells, active area, and materials. Power density higher than 1 kW/Kg and 1.5 kW/lt are reached by well designed stack with volume and weight lower than 10 lt/m^2 and 10 kg/m² respectively.

The electric behaviour of PEMFC can be evaluated by means of operating curves plotted in the (I,V) plane. The polarization curve, which shows the relationship between cell voltage and current density, is the index of the FC performances (Correa, *et al.*, 2001).

Assuming the gases ideal, the reagent partial pressures low, if compared with their critical pressures, and the operating temperature high, the output voltage of a single cell can be expressed as (Van der Merwe, *et al.*, 2002):

$$V_{FC} = E_{Nerst} - V_{att} - V_{ohm} - V_{conc}$$
(3)

where E_{Nerst} is the "thermodynamic potential" of the cell and represents its reversible voltage; V_{att} , the "activation overpotential", is the voltage drop due to the activation of the anodic and cathodic reactions and relates to the electrodes drop; Vohm, the "ohmic overpotential", is the ohmic voltage drop, measuring the ohmic losses associated with the conduction of protons in the solid electrolyte and internal electronic resistances; V_{conc}, the "concentration overpotential", is the voltage drop related to hydrogen and oxygen mass transport. The first term of (3) represents the FC operation open circuit voltage V_{oc} , whereas the others represent the reductions in the voltage due to operating conditions; finally VFC is the instantaneous cell voltage during load supply.

The reversible voltage is representative of the open circuit thermodynamic balance; it is a function of the operating temperature, the standard reference temperature (equal to 25 °C) and the pressure as:

$$E_{Nerst} = \frac{\Delta G}{2F} + \frac{\Delta S}{2F} \left(T - T_{ref}\right) + \frac{RT}{2F} \left[\ln\left(p_{H_2}^*\right) + \frac{1}{2}\ln\left(p_{O_2}^*\right)\right]$$
(4)

where ΔG is the free Gibbs energy change (J/mol), *F* is the Faraday constant (its value is 95,487 C), ΔS is the entropy change (J/mol), *R* is the universal constant of gases (8.314 J/K·mol), $p_{H_2}^*$ and $p_{O_2}^*$ are the partial pressure (atm) of the hydrogen and the oxygen respectively. *T* is the cell operating temperature (K) and T_{ref} the reference temperature. Using standard pressure and temperature for ΔG , ΔS and T_{ref} , (4) can be simplified to:

$$E_{Nerst} = 1.229 - 0.85 \cdot 10^{-3} [T - 298.15] + +4.31 \cdot 10^{-5} T \left[\ln \left(p_{H_2}^* \right) + \frac{1}{2} \ln \left(p_{O_2}^* \right) \right]$$
(5)

 V_{att} is connected to the speed of the reactions, which occur at electrodes, and depends on the energy possessed by the particles that collide. The least quantity of energy that the particles must possess, represents the activation energy. The activation overpotential for each electrode is calculated from the Buttler-Volmer equation (Correa, *et al.*, 2003). By considering only the dominant process, it is possible to express the activation overpotential as a function of the operating temperature, current and oxygen concentration at catalytic site [10]:

$$V_{act} = -\left[\xi_1 + \xi_2 \cdot T + \xi_3 \ln(c_{O_2}) + \xi_4 T \ln(i)\right] (6)$$
$$c_{O_2} = \frac{p_{O_2}^*}{5.08 \cdot 10^6 \exp(-498/T)}$$
(7)

where *i* is the cell operating current (A), c_{O_2} is the concentration of oxygen in the catalytic interface of the cathode (mol/cm³) and the ξ^{2} 's represent parametric coefficients for each cell model. These values are set in theoretical equations with kinetic, thermodynamic and electrochemical foundations. Coefficients may be determined by linear regression of experimental data, as reported in Table I (Amphlett, *et al.*, 1995; Yerramalla, *et al.*, 2003).

Table 1 Value of Parametric Coefficients

Parameter	Value
ξı	-9.44E-1
ξ2	3.54E-3
ξs	7.80E-5
ξ₄	-1.96E-4

The ohmic overpotential is a function of the resistance of both the electron transfer (in collecting plates and carbon electrodes) and the proton transfer (in the solid membrane). The equivalent resistance of the membrane can be calculated as (Laughton, 2002):

$$R_M = \frac{\rho_M l}{A} \tag{8}$$

In (8) ρ_M is the specific resistivity of the membrane ($\Omega \cdot \text{cm}$), *A* is the cell active area (cm²) and *l* is the thickness of the membrane (cm). The following numeric expression for the resistivity of Nafion membranes is usually used:

$$\rho_{M} = \frac{181.6 \left[1 + 0.03 \left(\frac{i}{A} \right) + 0.062 \left(\frac{T}{303} \right)^{2} \left(\frac{i}{A} \right)^{2.5} \right]}{\left[\lambda - 0.634 - 3 \left(\frac{i}{A} \right) \right] \exp \left[4.18 \cdot \left(\frac{T - 303}{T} \right) \right]}$$
(9)

where $181.6/(\lambda - 0.634)$ is the specific resistivity with no current at 30 °C; the exponential term in the denominator is the temperature factor correction if the cell is not at 30 °C and *T* is the absolute temperature of the cell (K). The parameter λ is a function of the relative humidity and stoichiometry relation of the anode gas. It can assume a value of about 14, under ideal conditions of 100% of relative humidity, and a value of about 22-23 in over saturated conditions. Using (8) for the membrane resistance, the following expression determines the ohmic overpotential:

$$V_{ohm} = (R_M + R_c) \cdot i \tag{10}$$

where R_c is the contact resistance, normally assumed equal to 0.3 m Ω .

The mass transport affects the concentrations of hydrogen and oxygen. This causes the decrease of the gasses partial pressures. Reduction in the partial pressure of oxygen and hydrogen depends on the electrical current and physical characteristic of the system. To express the voltage drop, it is possible to introduce the maximum current density, J_{max} , related to the max fuel speed rate under which the fuel is being used. Thus, the voltage drop due to the mass transport is:

$$V_{con} = -B \cdot T \ln \left(1 - \frac{J}{J_{\text{max}}} \right)$$
(11)

where B (V/K) is a constant depending on the cell and its operation state and J represents the actual current density of the cell. Usually it is possible to assume the values of such parameters as reported in Table II (Amphlett, *et al.*, 1996).

Table 2 Value of Parameters

Parameter	Value
В	5.3E-5 (V/K)
J_{max}	$0.5 (A/cm^2)$

The proposed final model has been performed. Assuming in the proposed model the cell thickness equal to 178 μ m and a value of λ equal to 23, the result of simulation is reported in Fig. 4. The polarization curve of a single cell, which focuses on the non linearity of the model, can be used to determine the static behaviour of a FC stack, defining and verifying the opportunities to couple in series or parallel configuration the cells in accordance with electrical load requirements.



Fig. 4. Simulated polarization curve

3. STACK OPTIMAL DESIGN

The opportunity of using a FC source as electrical generator is related to the possibility of designing an electrical source able to satisfy load requirements in terms of static and dynamic performances. At present, the reduced diffusion of the FC systems limits the scale economy and increases the importance of the design for determining a solution optimal in terms of cost reduction.

One of the problems that limits development and marketing on the large scale of fuel cell sources is the high manufacturing cost; this is tied up to the use of platinum electrocatalysts (see \$2), which defines the cell active surface area with a density of 0.4-0.5 mg/cm². To contain FC costs, according to the required performances in terms of efficiency and power, it is hence necessary to minimize the overall stack active area, taking into account the technological limits related to the realization of the stack.

The study of the FC physics behavior focuses on the evidence that the FC electrochemical generators are normally connected to the power system by an electronic converter, which operates to increase the FC voltage value to the electric system voltage. As a consequence, it would be interesting to investigate into FC performances in terms of different stack configurations (i.e. few cells of large area or a series of many cells of reduced active area), which directly affect the performance and active materials constituting the source. For the non linearity of the model the study can be performed by investigating into the characteristics by a series of analyses effected varying the area. On the basis of the electrochemical model above mentioned, the operational characteristics of stacks with different configurations have been derived, aiming to effect a comparative analysis for source optimal design. In the following the parameters reported in Table III are assumed.

Table 3 Used Data For Analyses

Parameter	Value
Minimum global area	$2000 (cm^2)$
Maximum global area	$3000 (cm^2)$
Temperature	333 (K)
Hydrogen pressure	1 (atm)
Air pressure	1 (atm)
J _{max}	$0.5 (A/cm^2)$
Max number of cells	50
Load losses	0.1%

In Fig.5 the representative curves of different stack configurations in terms of current-power are reported. Every curve represents the power provided by the stack as a function of the current feeding the load. It is possible to note that, for a fixed value of the active area (i.e. 2000 cm^2), the growth of number of cells in the stack (N) directly increases the maximum available power, but reduces, in the meantime, the applications in terms of operating current range. It can be summarized that stacks with high number of cells in series configuration are more suitable for the application with almost constant load. Assuming a fixed value of N (i.e. N=2), the output power grows, as expected, with the overall active area, as shown in Fig. 6, where the overall active area varies from 2000 cm² to 3000 cm² with step 200 cm^2 .

The examination of the curves shows how there are no differences for low operating currents, where the operating current density is less than the maximum cell current density. The curves are detached when the current grows and the J_{max} , being fixed, any



Fig.5. Output power varying N



Fig. 6. Output power varying overall active area

percent increase of overall active (in the figure equal to 10% of overall active area) determines the consequent linear growth of the maximum stack current.

These considerations characterize any value of N and, obviously, in absolute values, the increase of the maximum operating current between two areas is lower the higher the number of cells in series configuration. This is evident in Fig. 7, where the current-power diagrams are reported for different stack configurations in terms of cell number and overall active area. Every curve represents the power provided by the stack as a function of the current feeding the load, assuming the fixed number of cells in series in series configuration, N, and the fixed active area.

It is possible to note how the lower curve of N=3, which is representative of a global active area of 2000 cm², is above the upper curve representative of N=2 (global area of 3000 cm²) for about all the range of operating current. It results that, with a fixed global active area, the increase of N determines the substantial increase of the available output power.

For different configurations, voltage and current, which the power is provided with, vary; in particular, with a high value of N, the active area of a single cell is very little (i.e. for N=50 and active area=2000 cm² the single cell area is only 40 cm²) and, because the maximum current density cannot be varied, the stack current is very reduced. In Fig.8 the peak value of the available power is reported for different stack configurations. The curves, parameterized as a function of overall active area, show as the growth of the number of cells increases the available maximum output power; it is focused on the evidence that, according to the extreme splitting up of the global active area, the slope of the curves reduces increasing the numbers of cells; so that, when the high power is required with high voltage is not sufficient to increase N but it is also necessary to increase the overall active area involving inevitably



Fig. 7. Output power for different stack configurations

the growth of the source cost.

For every fixed value of global active area, an optimal value of N can be defined. N_{opt} index can be determined as the number of cells in series which is not convenient to overcome because the increase of N involves an increase in costs without determining a significant increase in the output power, due to the effects of the load losses.

For instance in Fig. 9 it is represented the maximum power curve, when a global active area equal to 2000 cm^2 and a maximum number of cells of 700 are considered.

The N_{opt} could be determined as the representative of the maximum available power of the stack, for an assigned value of the global active area without take into account any constraint on operating voltage.

To suit the FC source to the converter requirements, it is normally necessary to reduce the voltage variation around its nominal value. Therefore it is however necessary also to determine how the output power of the stack varies, in different configurations, when the voltage varies in this range. Assuming a cell voltage basic value equal to 0.6 V, which is a mean operating value of the cell, and a voltage drop of 10%, caused by the growth of load current, it is possible to determine the variation of the output power, in percentage of the maximum output power, for the different stack configurations. The results are shown in Fig. 10 where the configuration with N < 10are neglected; it is evident that the variation of $\Delta P\%$ decreases as N grows. In fact, the cell voltage being fixed, with the increase of N the cell operating point approaches the peak of the output power curve (see Fig. 6); in this zone the curve has a low gradient so the variation of cell voltage determines a smaller increase in the output power. The curve also indicates that the stack configurations with a high value of N if on one hand determine an increase in the maximum output power, on the other have low overload capability because, the stack voltage having to remain within converter requirements, the permitted variation of ΔP is only a few percent. Instead, for stack configurations with low N the output power can vary by 25% remaining the stack voltage in the limits dictated by the converter.

4. CONCLUSIONS

In the paper there are investigated on the operation characteristics of a polymeric electrolyte membrane (PEM) FC, suitable for the application in electric



Fig. 8. Maximum output power for different stack configurations



Fig. 9. Maximum output power varying number of cells in the stack



Fig. 10. Variation of $\Delta P\%$ for a variation of V_{output}

power systems. To characterize the FC electrical behavior, the steady-state electrochemical model is defined and the incidence of the stack configuration on the FC system performances are examined.

The results of the analyses focus on a series of considerations, which relate different stack design to the available electrical performances. The analyses are addressed to examine both the performances of the stack in terms of maximum available energy and power as function of an optimal number of series cells and the integration of the FC source with the dedicated converter. The main result show as the growth of the number of cells increases the available maximum output power, but makes the stack particularly suitable for the application with constant load. In particular, with reference to the converter integration, stacks characterized by a reduced number of cells in series configuration result more able to limit the voltage variation with varying loads, favoring the interaction of the FC source and the converter for different values of output power.

ACKNOWLEDGMENTS

The authors would like to acknowledge G. Paparo

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