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Polymer electrolyte fuel cell stack research and development

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ABSTRACT

The research activity in polymer electrolyte fuel cell (PEFC) is oriented to the evolution of components and devices for the temperature range from 20 to 130 °C, and covers all the aspects of this matter: membranes and electrodes, fuel cell stack engineering (design and manufacturing) and characterization, computational modelling and small demonstration systems prototyping. Particular attention is devoted to portable and automotive application. Membranes research is focused on thermostable polymers (polyetheretherketone, polysulphone, etc.) and composite membranes able to operate at higher temperature (>100 °C) and lower humidification than the commercial Nafion®, while Pt load reduction and gas diffusion layer improvement are the main goals for the electrode development.

PEFC stack engineering and characterization activity involve different aspects such as the investigation of new materials for stack components, fuel cell modelling and performance optimization by computational techniques, single cell and stack electrochemical characterization, development of investigation tools for stack monitoring and data acquisition. A lot of work has been focused to the fuel cell stack architecture, assembling, gas leakage and cross-over reduction (gasketing), flow field and manifold design. Computational fluid dynamics studies have been performed to investigate and improve reactants distribution inside the cell. A flow field design methodology, developed in this framework and related to serpentine like flow field, is actually under investigation.

All of these aspects of PEFC stack research are realized in the framework of National and European research projects, or in collaboration with industries and other research centres.

In the present work our stack research activity is reported and the most important results are also considered.

1. Introduction

Polymer electrolyte fuel cells (PEFCs) are becoming an interesting power source in different application fields, from small electronic portable units to medium size stationary systems. This wide range in power output implies a great variation of fuel cell operative conditions, active area size and membrane electrode assembly (MEA) properties. Consequently, the improvement of PEFC stack research involves all the components, from the membrane to stack auxiliaries with special attention to materials, hardware design (channels geometry, manifold, sealing, etc.) and fuel cell components coupling. These aspects have been deeply discussed and investigated in several scientific works [1–7]. Furthermore,

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Nomenclature[1]

RC	Reference cell
α_1	calculated and RC channel width ratio
α_2	calculated and RC number of parallel channels ratio
β_1	calculated and RC active area shape factor (or aspect) ratio
β_2	calculated and RC covering factor ratio
γ_1	calculated and RC pressure drop ratio
γ_2	calculated and RC gas velocity ratio
γ_3	calculated and RC Reynolds number ratio

recent advances in PEFC technology have opened the road to commercialization, and the system simplicity, weight and volume reduction, become key aspects for developers. As a result, "air breathing" or "open cathode" stack configurations, operating at low humidity or completely dry gases, are emerging as such classes of PEFC stacks for power devices actually supplied by conventional batteries.

Furthermore, research efforts are addressed to cost reduction in terms of new material development (catalyst, membranes, hardware materials) and components design [1,4,6,8–13].

Looking at the open questions, the PEFC research activity has been directed both to the acquisition of the know how on single component development, and the whole system survey with special regards to the interaction of components.

Despite many studies about the influence of different parameters on the stack performance and efficiency [3,14–17], till today a standard methodology to approach the stack design is not available. Thus, a stack engineering activity, supported by numerical analysis, has started since 2001 involving the following subjects:

- dimensioning methodology studies;
- modelling: CFD (fluid dynamic), FEM (mechanical simulation) studies;
- stack and system design; and
- characterization tests on small size PEFC stack.

2. Dimensioning and methodology studies

Fuel cell design and optimization are generally carried out in small size cell; nevertheless, in practical applications an increment of active area is often required. However, if the fuel cell geometry is increased, the reactant distribution complexity makes difficult the performance maintenance. This problem has been approached by searching a set of dimensioning criteria able to define the new flow field geometry, and this criteria has been used to evolve a software named "*FDES*" for flow field selection [18]. The "*FDES*" software uses an algorithm to generate all the flow-field designs allowed by imposed geometrical constrains, and select that one matching the fluid dynamic target. To validate this method it was applied to a sample case by scaling up a single serpentine 5 cm^2 active area (AMEA) lab cell up to 125 cm^2. Factors related to number and width of channels (α_i), MEA design (β_i), pressure drop and fluid dynamic aspects (γ_i) were considered to evaluate the procedure sensibility to the inputs. The evolved algorithm generates and selects the flow channels design (single serpentine, ribbed-serpentine or multiple serpentine) that matches the requested geometrical and fluid

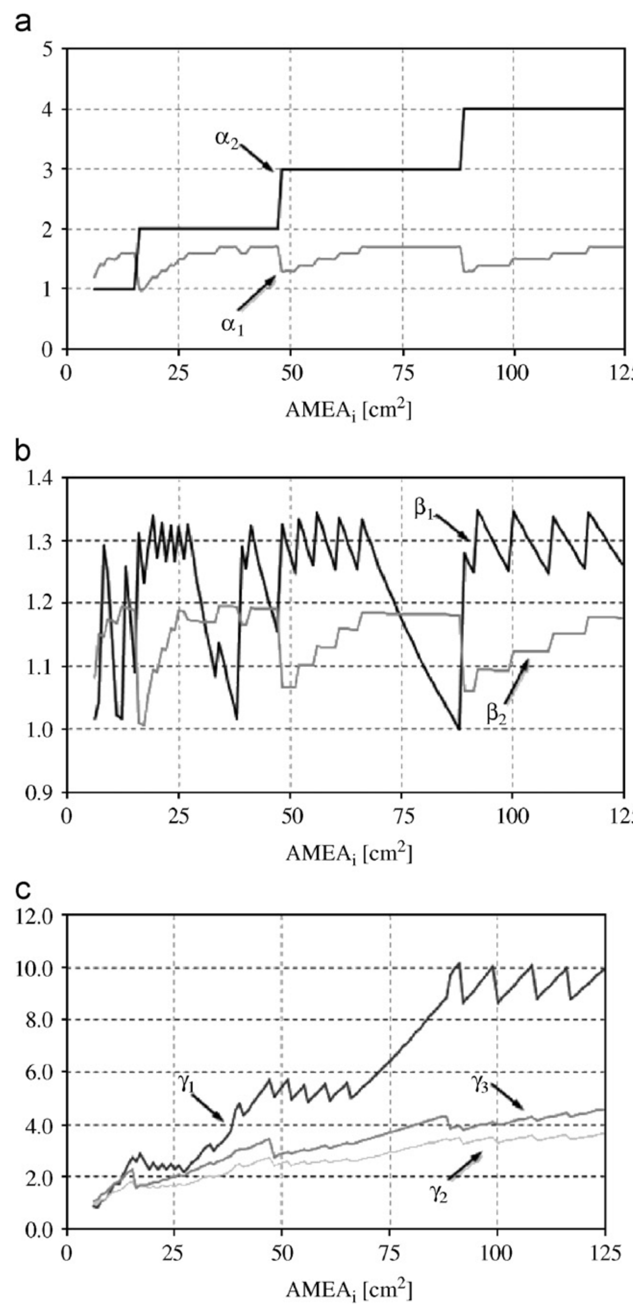

Fig. 1 – Scale factor profiles as a function of the active area cell: (a) α_1, α_2; (b) β_1, β_2; (c) γ_1, γ_2, γ_3.

[1] (see Ref. [20] also).

Table 1 – Inputs data for selection algorithm, the outputs are plotted in Fig. 1

Parameter name	Parameter meaning	Ref. cell value	Screening range
AMEA	Cell active area (cm^2)	5	6–125
LCAN	Channel width (mm)	1.0	1–2.5
HCAN	Channel depth (mm)	1.0	1.0
LCOS	Rib width (mm)	1.0	1.0
NDIV	Number of parallel sub-channels	1	1–5
N	Switch back number of serpentine	11	11–13–15
χ	Covering factor (contact surface to cell active area ratio)	0.54	0.50–0.65
ζ	Active area aspect ratio	0.88	0.88–1.19
Δp	Pressure drop (mbar)	9.60	–
Re	Reynolds number	82.64	–
v_{med}	Mean inlet velocity (m/s)	3.32	–

dynamics constrains. Fig. 1 shows the results obtained for a serpentine geometry and referred to the data inputs listed in Table 1.

Fig. 1(a) shows the behaviour of parameters α_1 and α_2 related to channel width and to the number of parallel sub-channels in each serpentine as a function of AMEA respectively. The sub-channels number (α_2) increases from 1 to 2, 3 up to 5 by increasing the active area; it means that, when a large area is requested, the number of parallel sub-channels has to be increased. At the same time, the channels width (α_1) increases for each constant α_2 value, and shows a periodic trend due to α_2 discontinuities. Stepwise behaviour is related to the finite increment imposed scanning LCAN screening range, In some cases, α_1 remains unvaried because it is limited by the imposed value of serpentine switch back number ($N-1$) that increases with AMEA.

As a consequence of these results, the simple serpentine can be considered for small active area less than 15 cm^2, and the ribbed or multiple serpentine flow field can be considered for greater active area.

Fig. 1(b) describes the behaviour of parameters β_1 and β_2 related to active area aspect ratio and covering factor, respectively.

The β_1 that ranges from 1.0 to 1.35 (deriving from the ratio between reference cell value and screening range in Table 1) is correlated to the sub-channel width (α_1); therefore, if we consider the stepwise trend of α_1 for each α_2 in Fig. 1(a), when α_1 and AMEA increase β_1 increases too, while if α_1 is constant and AMEA increases β_1 decreases and approaches the reference cell active area aspect ratio.

This behaviour determines the zig-zag trend. On the other hand, because β_2 is directly correlated to the open area deriving from the sub-channels width it maintains the same trend of α_1.

Fig. 1(c) shows the results obtained for $\gamma_1, \gamma_2, \gamma_3$ parameters that are related to the fluid dynamic aspects of the flow field. Notice that, increasing AMEA the pressure drop parameter (γ_1) increases if α_1 is constant and N (the serpentine length) increases.

On the contrary, when N is constant and α_1 increases, the pressure drop decreases determining the zig-zag trend. In the same way, the inlet gas velocity parameter (γ_2) and the Reynolds number parameter (γ_3) increase.

Following these results it is possible to define the scaled flow field geometry and design.

3. Modelling: CFD (fluid dynamic), FEM (mechanical simulation) studies

The ability of numerical models for predicting fuel cells behaviours has been proved in the last years [6,8,14,15,19,20]. Particularly, CFD and FEM studies have been used as a useful tool for optimizing components and investigating local phenomena inside the fuel cell. CFD has been used to evaluate local distribution of fluid dynamic variables such as velocity, pressure and temperature in the fuel cell flow field and estimate the interaction of the flow path and the porous electrode. In Fig. 2 an example of the CFD analysis results for gas velocity and pressure distribution at cathode side for a five channel serpentine flow field is reported.

Studies of different flow field configurations (ribbed, serpentine, multiple serpentine and interdigited) have pointed out the relevance of flow field and porous electrode interaction [18]. In particular, the presence of a secondary flow, termed cross flow also, has been highlighted in the serpentine like flow fields, where a fraction of the total mass flow rate is driven, from a channel to the adjacent one through the porous backing of the electrode due to the pressure difference. Availing of CFD, this phenomenon has been quantified introducing a coefficient ψ, defined as the ratio of total inlet mass flow rate in the fuel cell and mass flow rate passing through the porous layer. This coefficient is related to the ability of the flow field to provide reactants to the electrode. It is expected that this kind of approach could be used to design the fuel cell flow field.

However, more detailed studies on the local distribution of electrochemical quantities such as current density on the MEA surface, water activity and conductivity of the membrane, over potential losses and species concentration have been recently performed by the use of a commercial code es-PEFC from CD Adapco [6,15]. Further analysis of temperature distribution on the bipolar plates has been conducted to design the cooling system of single cell and stacks.

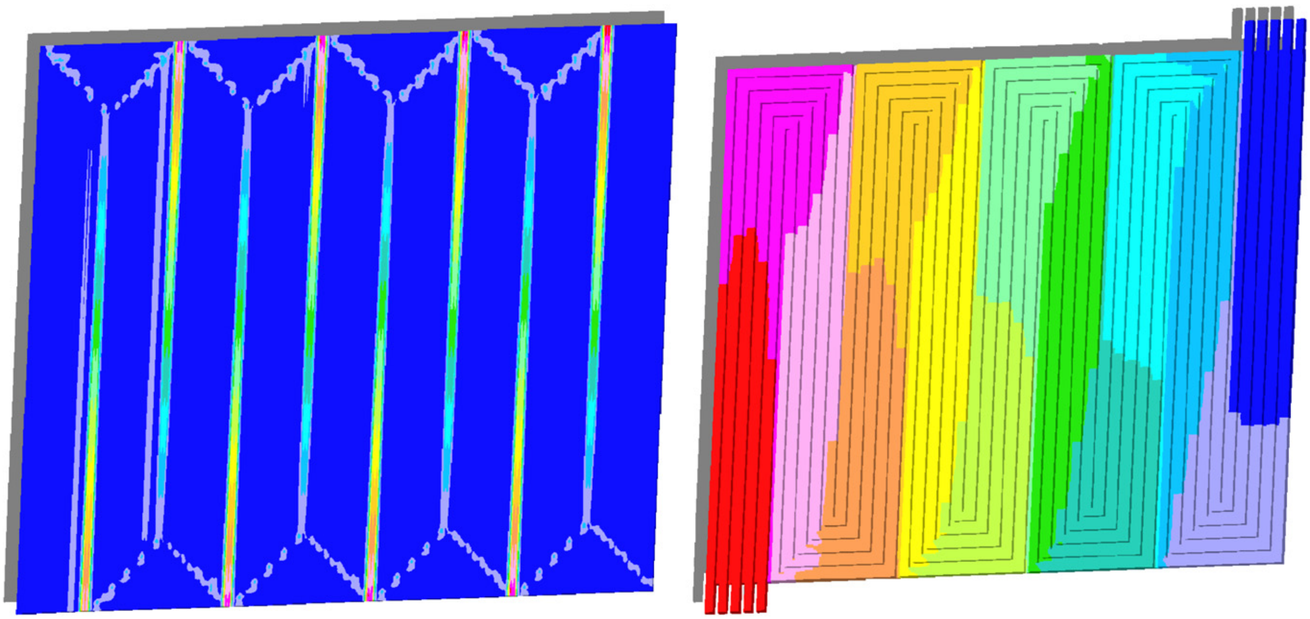

Fig. 2 – Example of CFD analysis results plot on a multiple serpentine flow field; velocity distribution (left) and pressure distribution (right) along the channel maps.

Stress and strain analysis of fuel cell hardware parts have been also conducted using the De Saint Venant theory and FEM analysis. Numerical models of graphite bipolar plates have been realized to verify stresses distribution over the bipolar plate and prevent mechanical failure. In fact, the presence of holes for manifold allocation and ribs of the flow channels yields to local over stresses of materials that are not determinable by the classical theory. Clamping systems have been also investigated to make uniform distribution of contact pressure between bipolar plates and MEA. This aspect is important for assuring a good electrical contact between conductive plates and surface electrode and prevent gas leakage. Experimental evidence of the contact pressure between gaskets and bipolar plates has been also fulfilled by the use of pressure sensitive sheets.

4. Fuel cell and stack design

Fuel cell and stack design engineering activity has the aim to improve the performance of devices in terms of power density and specific power. Single cell design is generally realized on the basis of experimental purposes. Stack design is more complicated because power and overall voltage target must be taken into account and MEA loss of performances in respect to the lab cell must be minimized. Moreover the stack final application, specific geometrical requests and cooling system must be considered also.

The first step of a fuel cell design is the definition of specifications to be accomplished, namely the power and voltage and subsequently the total current. On the basis of the electrode polarization curve, the current density and single cell voltage could be selected; therefore, the number of the cell and the total active surface area is determined as a consequence. As electrode operative conditions, such as anodic and cathodic reactants temperature, pressure and humidity, are known, reactant mass flow rate and thermo-physical properties of the stack inlet streams can be calculated. Once the active surface area shape factor is selected, the flow field parameters (channel height and width, rib width, flow path length, open ratio) and typology (serpentine, ribbed-serpentine and multiple serpentine) could be fixed by using the "*FDES*" software described before [18] that selects the flow field having the desired pressure drop. The bipolar plate layout is defined when manifolds size and position with respect to the flow field area are chosen. Total mass flow rate and pressure distribution in manifolds have to be considered for the manifold cross-sectional area selection. In fact, an underestimation of manifold cross-sectional area and flow field inlet cross-sectional area ratio can lead to a reactant mass maldistribution inside single cell of the stack. Otherwise overestimating the manifold cross-sectional area ratio, reduce the specific power density of the fuel cell in terms of active area to total area of the plate.

Stack cooling is another important aspect of the fuel cell design because a correct thermal management is necessary to assure cell stable performance. The definition of cooling fluid, air or water, is fundamental to accomplish the bipolar plate design (architecture, gasketing and so on) and depends on fuel cell application. Experience in this field has been gained by designing several small stacks and realizing some of them.

Reference materials used for stacks realization are the following:

- gas diffusion electrodes (GDE) [9–11,21] handmade by a spray technique;
- commercial Nafion membranes;
- composite graphite plates;

- current collectors realized by copper and protected against the oxidation by interposing a flexible graphite sheet between the copper and bipolar plates; and
- end plates of aluminium alloy.

Starting from these components a 100 We PEFC stack operating at low pressure (0.5 barg) with humidified hydrogen and air was realized [22]. It is composed of 10 cells with an active surface area of 49 cm^2, and it can operate between 5 and 7 V with a total current load in the range of 10–25 A.

A serpentine-ribbed flow field was adopted as reactant distributor for the single cell. The cooling of the unit was assured by 6 W blower that forces air inside cooling fins obtained by alternating bipolar plates with different area. A power of 105 W at about 5 V that corresponds to a power density of about 210 $mW\,cm^{-2}$ was obtained at 80 °C with a stoichiometric ratio 1.5/4 and 75/80% relative humidity (RH) for H_2 and air, respectively.

Successively, a hydrogen fed, air breathing stack of 15 W has been designed and realized for portable application [23]. The unit does not require forced air to be fed at the cathode because it is able to spontaneously breath air from the surrounding. Hydrogen is supplied in end off configuration at a pressure of 0.2 barg. The stack is composed of 10 cells with an active surface of 25 cm^2 and generates 15 W (rated power) at 7 V with a maximum power 21 W at 4.3 V. This stack has been used as the core of a demonstration mini-power unit able to supply to a portable DVD player [24]. A stabilized power output was reached by a homemade DC–DC converter having an output of 11.3 V. A 40 hydrogen litres metal hydride tank (provided by solid H) with cooling fins was used for hydrogen storage.

The realized devices are reported in Fig. 3.

5. Characterization tests on small size PEFC stack

The research activity on stack concerns electrochemical and mechanical tests also, aimed at confirming the modelling results, design reliability and finding practical solution for system manufacturing. Electrochemical tests are carried out on a homemade test station that controls single cell and stack operative conditions by means of a proprietary software. A computer data acquition system is able to automatically perform the electrochemical characterization of the unit, such as polarization curve, time test at constant current or voltage and cyclic tests.

A conditioning period of the cell, in which the fuel cell is fed with humidified gases at the working temperature and the cell temperature increased up to set point is performed before the test start up. After this procedure, the selected test can be fulfilled automatically.

Several studies on PEFC from 1 to 10 cells have been carried out, namely:

- influence of RH of gases on the performance and stability of the cells;
- influence of the different flow rates of hydrogen and air reactants;
- influence of different cooling system approaches on cell temperature and performance parameters;
- pressure drop measurements under and out of work, in order to validate CFD studies and to diagnose either flooding or drying conditions inside the stack;
- stability tests to verify the life time of the components and the resistance to stress in work conditions;
- mechanical test of stack components to verify that sealing, gas distribution and clamping system are functional and reliable.

6. Conclusions

As illustrated above, continuous R&D of polymer electrolyte fuel cell stacks is being carried out leading to small stack and systems evolution, to the realization of a stack engineering methodology and to experimentally confirm the modelling studies.

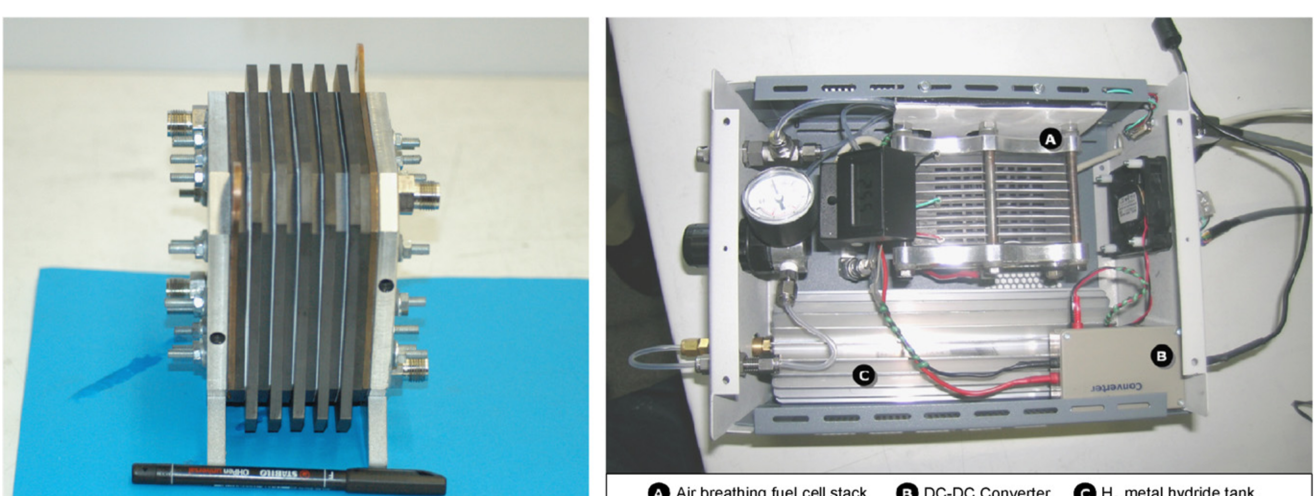

Fig. 3 – Frontal view of the 100 W stack (left), top view of the MPU system with the air breathing stack (A) and DC–DC converter (B).

All these experiences will be the starting point of future research activities. These activities foresee the development of a 250–500 W stack operating at 90–120 °C; the realization of a 0.5–1 kW air cooled stack for portable applications and the realization of a 7 kW liquid cooled stack for stationary applications.

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