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Influence of the operational parameters on the performance of polymer electrolyte membrane fuel cells with different flow fields

Abstract The need for more efficient energy conversion devices that can operate with a minimum of environmental pollution has drawn much attention to fuel cells in recent years. However, the performance of low-temperature fuel cells, like the polymer electrolyte membrane fuel cell (PEMFC), needs substantial improvement. This work describes the effect of the operational and structural parameters on PEMFCs with different flow fields and gas diffusion electrodes built with conventional procedures. The results show that, under particular conditions, significant improvements in performance can be obtained by using interdigitated flow fields of adequate dimensions.

Introduction

The depletion of fossil fuels and the degradation of the environment are key issues in modern society. There is an urgent need to substitute fossil fuels by renewable fuels and at the same time find new energy conversion methods that preserve the environment, particularly in densely populated urban centres. Because of these problems, much effort has been devoted in recent years to the development of different types of fuel cells [1, 2]. Of these, the polymer electrolyte membrane fuel cell (PEMFC) has attracted much attention. Working with hydrogen and oxygen (from the ambient air) the

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PEMFC presents a high power density, which makes it ideal for transportation applications and portable systems. Furthermore, the PEMFC can operate by oxidizing a liquid fuel, like methanol, directly, which makes it convenient for applications where the problems of production, storage and distribution of hydrogen are difficult to overcome. However, the power densities that can be achieved with methanol are still too low when compared to those obtained with hydrogen.

The PEMFC has a simple design. The gas diffusion electrodes are usually hot-pressed onto a proton exchange membrane to fabricate the membrane and electrode assembly (MEA), which is placed between two electrically conducting plates that act as current collectors and the reactant gas distribution network.

Presently, several problems limit the performance of the PEMFC. Neglecting a possible contamination of hydrogen with CO, if it is produced by reforming, the main ones are connected with the oxygen reduction reaction. At low current densities the activation overpotential of this reaction is large, and at high current densities other problems may dominate, like the transport of oxygen to the catalyst sites, which is often made more difficult if, due to the temperature of operation of about 80 °C, liquid water is present at the cathode, leading to a two-phase flow of oxygen gas to the catalyst sites. Actually, water management is one of the key issues in PEMFCs and has been treated with experimental [3] and modelistic [4] approaches.

Several types of gas diffusion electrode are employed in PEMFCs. Usually they are constituted by a gas diffusion layer and a catalytic layer made of a suitable conducting support like carbon cloth or paper [5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18]. The diffusion layer of the electrodes sits on a reactant gas flow field that can be machined or moulded onto the current collector. The more simple flow field is constituted by parallel channels. Alternatively, a serpentine-type flow field, which may have a single, double or triple channel, promotes a longer contact between the reactant gas and the backing of the gas diffusion electrode and is preferred in many

designs. More recently, still another flow field type, the interdigitated, was found to increase the performance of the cell and contribute to expel liquid water from the electrode [19, 20, 21]. For a better understanding of the problem, the three flow field patterns are depicted schematically in Fig. 1. In the interdigitated flow field, the inlet and outlet channels are not directly connected and the reactant gas is forced by convection to pass through the backing layer of the electrode. This contrasts with the parallel channel system in which the reactant gas diffuses into the backing layer of the electrode. In the serpentine type the main transport mechanism is diffusion but, if a substantial pressure drop between inlet and outlet exists, some convection may also contribute.

A comparison between different flow fields has shown improved performances with the interdigitated, but the comparison has not been made under sufficiently different operational conditions of the PEMFCs. In this work, the performance of PEMFCs with parallel, serpentine and interdigitated flow fields of varying dimensions are compared under different operational conditions and different compositions of the gas diffusion electrodes. In this last case, the influence of some structural parameters of the electrodes, like the thickness and the quantity of PTFE in the diffusion layer and the amount of Nafion in the catalytic layer of the electrode, are considered.

Although for practical applications the objective is to develop PEMFCs that work with ambient air, the use of pure oxygen in the cathode may be an important addition to our knowledge of the processes that take place in the PEMFC. Because of this, the performance of the PEMFC was studied here first with oxygen and then with air.

Experimental

Gas diffusion electrodes were made by a combined filtration/rolling procedure. The diffusion layer was made by applying a mixture of carbon powder (Vulcan XC-72, Cabot) and a PTFE suspension (T-30, DuPont) onto a carbon cloth (Stackpole, PWB-3, 350 μm). On top of the diffusion layer, a catalytic ink made with carbon powder, Pt catalyst (20% Pt/C E-TEK) and Nafion solution (5% Aldrich, H^+ form) was applied. This ink was made in isopropanol, to break the hydrophobicity of the diffusion layer. Before use, the carbon cloth was treated in an inert atmosphere at 450 °C for 1 h and washed with hot HNO_3 solution.
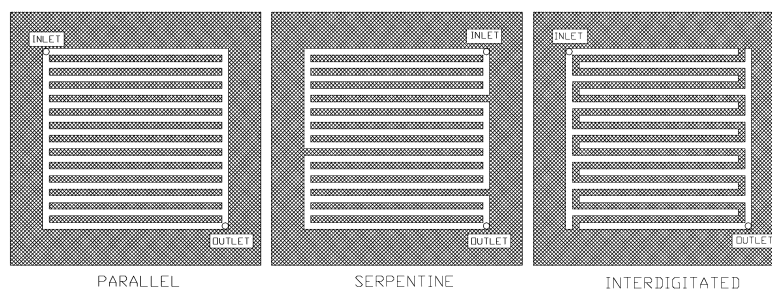
The electrolytes were 115 Nafion membranes (DuPont, 125 μm). The membranes were treated with 3% H_2O_2 for 1 h at 80 °C followed by treatment with a 0.5 mol L^{-1} H_2SO_4 solution.

The electrodes were made with different compositions of the diffusion and catalytic layers, and the main parameters are given in Table 1. The different electrodes were all tested with the different flow fields described below.

The MEAs were made by pressing two electrodes onto a membrane at 125 °C and 5 MPa for 2 min.

Single cells were mounted with gas diffusion electrodes having a geometric area of 4.6 cm^2. The single cells were clamped between aluminium blocks, and it was necessary to determine the torque that should be applied to the bolts. A torque of 20 lb in resulted in a low performance and leaks when the cell was under pressure. Increasing the torque to 30 lb in, the cell performance was as expected and no leaks were detected. A further increase to 40 lb in did not improve the performance and there was some risk of damaging the electrode. Therefore, all cells were assembled with a final torque of 30 lb in.

Before entering the cell, the reactant gases were humidified by passing through water contained in temperature-controlled metal bottles. The operational parameters of the cell, the polarization curves and the data recording were managed by a controller station made in-house [22]. Except where indicated, the single PEMFCs were operated at 80 °C, with oxygen or air humidified at 85 °C [$T(O_2)$ or $T(air)$] and hydrogen humidified at 95 °C [$T(H_2)$]. The single cells operated with either hydrogen/oxygen or hydrogen/air with different gas flows identified by a parameter λ, where $\lambda = 1$ indicates stoichiometric flow at 2 A cm^{-2}. In all cases the flow of hydrogen corresponded to $\lambda = 2$ for a current density of 2 A cm^{-2}.

The reactant gas flow fields were machined into carbon plates (Carbono Lorena, HCB5/2F). The flow field designs used were the parallel, serpentine and interdigitated channels and the characteristics of the different flow fields used are given in Table 2. Four different interdigitated flow fields were used, three of them with different dimensions of the depth, height and channel separation, labeled A, B and C. The design labeled A2 has the same dimensions as that labeled A but incorporates one more channel. No particular problems were detected in the machining of the interdigitated pattern or problems of fragility when the bipolar plate was clamped in the cell. Therefore, it is expected that in the scale-up of interdigitated bipolar plates the same considerations as in plates with the traditional patterns will have to be made.

Results and discussion

Hydrogen/oxygen PEMFCs

The experiments with pure oxygen were directed to the examination of the effect of the electrode structure on performance, by using different compositions of the diffusion and catalytic layers of the electrode, shown in Table 1. Modifying the composition of the electrode in terms of catalyst load and Nafion content, the properties

Fig. 1 Schematic diagrams of the three flow-field patterns

PARALLEL   SERPENTINE   INTERDIGITATED

Table 1 Compositions of the diffusion and catalytic layers of the electrodes

Electrode	Diffusion layer		Catalytic layer	
	PTFE (% w/w)	C + PTFE (mg cm^{-2})	Nafion (% w/w)	Catalyst (mg cm^{-2})
1	15	6.0	31.0	0.1
2	15	6.0	35.5	0.1
3	15	6.0	31.0	0.4
4	15	6.0	35.5	0.4
5	15	6.0	40.0	1.0
6	10	4.0	35.5	0.4
7	10	7.5	31.0	0.4
8	15	6.0	35.5	0.4
9	15	6.0	31.0	0.4

Table 2 Characteristics of the different flow fields

Flow field design	Channel height (mm)	Channel width (mm)	Channel separation (mm)	Number of channels (inlet/outlet)	Contact area gas/electrode (mm^2)
Parallel	0.8	0.8	0.8	14	314
Serpentine	0.8	0.8	0.8	14	316
Interdigitated A2	0.8	0.8	0.8	7/7	255
Interdigitated A	0.8	0.8	0.8	7/6	226
Interdigitated B	1.4	1.4	1.4	4/3	227
Interdigitated C	2.0	2.0	2.0	3/2	236

of the triple phase catalyst/electrolyte/gas, where the electrochemical reactions take place, can be modified. In these experiments the flow of oxygen, supplied at atmospheric pressure, corresponded to $\lambda = 3$ for a current density of 2.5 A cm^{-2}.

In all the experiments with oxygen, the interdigitated flow field showed a better performance than the conventional systems. The design called interdigitated A was in general better than the serpentine design, and this was followed in performance by the interdigitated designs called B and C (Table 2). Some exceptions were observed in favour of the interdigitated patterns. For example, the cell voltage-current density curves in Fig. 2 for electrode 1 show a better performance for all the interdigitated designs as compared to the serpentine. Electrode 1 has a low platinum load and the catalytic layer is particularly thin. The results may therefore be interpreted as indicating that the convective flow promoted by the interdigitated flow fields allows a better utilization of the catalyst. The comparative results can also be observed in Table 3, which shows the cell voltage at the maximum power density and the corresponding current densities for the different electrodes (electrodes 1–7) and flow fields. From the data in Table 3 it can be calculated that the gain in performance in going from the serpentine to the interdigitated A design is 28%. An increase in the content of Nafion in the electrode (electrode 2, Table 1) shows an increase in performance for all the systems, indicating a better contact between the catalyst and the electrolyte. This is not a general rule, because a too high increase in Nafion content may worsen the performance. Electrodes 3 and 4, whose behaviour is illustrated in Fig. 3 for electrode 3, show an
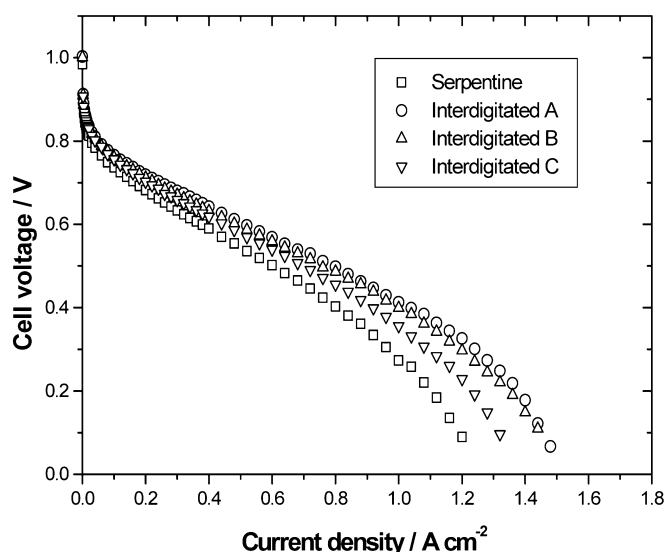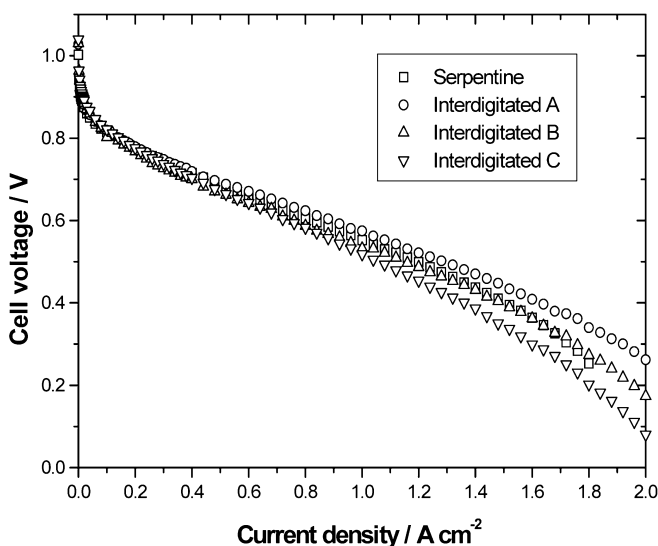

Fig. 2 Cell voltage-current density curves for a single H$_2$/O$_2$ (at 0.1 MPa) PEMFC at 80 °C with different flow fields and electrode 1. $T(\mathrm{H_2}) = 95$ °C, $T(\mathrm{O_2}) = 85$ °C, flow of hydrogen $\lambda = 2$ at 2 A cm^{-2}, flow of oxygen $\lambda = 3$ at 2.5 A cm^{-2}; Nafion 115 membrane

increase in performance because they have a higher platinum load. However, electrode 3, with a lower Nafion content, shows a better performance than electrode 4. This is because a moderate increase in the amount of Nafion improves the catalyst utilization, but a too high Nafion content represents a barrier to the transport of reactant gas to the active sites. The results for electrode 5 (Table 3) show that an excess of Nafion
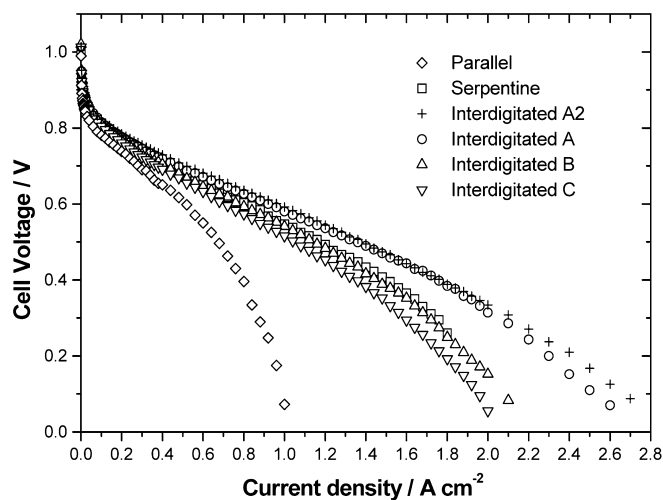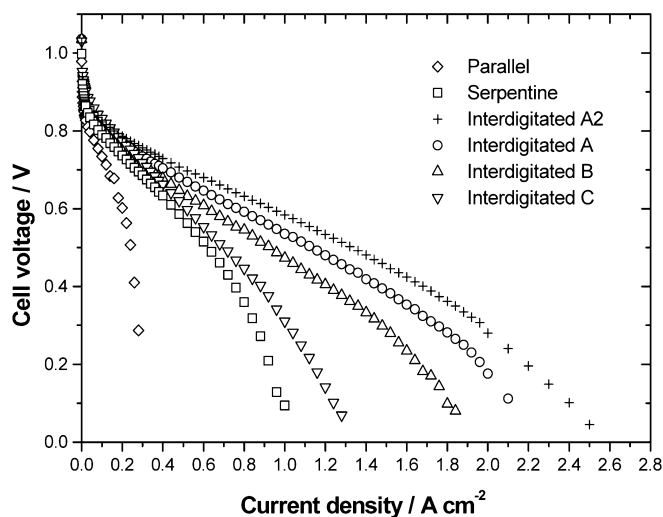
Table 3 Cell voltage at maximum power densities and corresponding current densities; electrodes 1–7 and different flow fields for a H_2/O_2 PEMFC

Flow field design	Electrode 1		Electrode 2		Electrode 3		Electrode 4		Electrode 5		Electrode 6		Electrode 7	
	V	A cm^{-2}	V	A cm^{-2}	V	A cm^{-2}	V	A cm^{-2}	V	A cm^{-2}	V	A cm^{-2}	V	A cm^{-2}
Parallel	–	–	–	–	–	–	–	–	–	–	0.496	0.68	0.563	0.22
Serpentine	0.403	0.80	0.394	1.20	0.437	1.40	0.420	1.40	0.376	0.84	0.447	1.36	0.461	0.68
Interdigitated A2	–	–	–	–	–	–	–	–	–	–	0.432	1.64	0.424	1.60
Interdigitated A	0.384	1.08	0.398	1.24	0.448	1.48	0.383	1.64	0.390	0.96	0.429	1.64	0.435	1.36
Interdigitated B	0.417	0.96	0.406	1.08	0.431	1.40	0.380	1.48	0.431	0.38	0.434	1.36	0.405	1.20
Interdigitated C	0.418	0.88	0.409	0.92	0.413	1.32	0.394	1.08	–	–	0.437	1.24	0.446	0.80

Fig. 3 Cell voltage-current density curves for a single H_2/O_2 (at 0.1 MPa) PEMFC at 80 °C with different flow fields and electrode 3. $T(H_2) = 95$ °C, $T(O_2) = 85$ °C, flow of hydrogen $\lambda = 2$ at 2 A cm^{-2}, flow of oxygen $\lambda = 3$ at 2.5 A cm^{-2}; Nafion 115 membrane

Fig. 4 Cell voltage-current density curves for a single H_2/O_2 (at 0.1 MPa) PEMFC at 80 °C with different flow fields and electrode 6. $T(H_2) = 95$ °C, $T(O_2) = 85$ °C, flow of hydrogen $\lambda = 2$ at 2 A cm^{-2}, flow of oxygen $\lambda = 3$ at 2.5 A cm^{-2}; Nafion 115 membrane

in the catalyst layer reduces the performance drastically. The convective flow of the interdigitated flow field is not efficient enough to reach current densities comparable to the previous cases. Nonetheless, the maximum power density is about 20% higher than that of the cell with a serpentine flow field.

The effect of the composition of the diffusion layer of the electrode was studied with electrodes 6 and 7 (Table 1). The cell voltage-current density curves for a PEMFC operating with electrode 6 are shown in Fig. 4 and those with electrode 7 in Fig. 5, for the different flow fields. The results in terms of the cell voltage at maximum power density are presented in Table 3. The effect of increasing the diffusion layer thickness depends strongly on the type of flow field used. The most efficient system, that with the interdigitated A2 flow field, shows little sensitivity to the thickness of the diffusion layer, with a decrease in the maximum power density of about 5%. The same can be said for the interdigitated A and B designs, although these show much lower performance. On the other hand, the decrease in performance for systems with the parallel, serpentine or interedigitated C

Fig. 5 Cell voltage-current density curves for a single H_2/O_2 (at 0.1 MPa) PEMFC at 80 °C with different flow fields and electrode 7. $T(H_2) = 95$ °C, $T(O_2) = 85$ °C, flow of hydrogen $\lambda = 2$ at 2 A cm^{-2}, flow of oxygen $\lambda = 3$ at 2.5 A cm^{-2}; Nafion 115 membrane

designs is very large, reaching almost 100% for the serpentine flow field.

The influence of the pressure drop (measured as the difference in pressure between gas input and output of the cell) on the operating conditions was measured in cells assembled with the different flow field plates, using electrodes similar to electrode 3, and the results are presented in Table 4.

According to Table 4, the pressure difference for the interdigitated type A is much smaller than for the types B and C. This is probably due to the larger number of channels of the interdigitated A, which promotes a better distribution of reagent on the electrode, and this leads to a better efficiency. Thus, the number of channels, and their dimensions, in an interdigitated pattern is an important factor in determining the homogeneity in the distribution of the reactant.

Hydrogen/air PEMFC

In the hydrogen/air cell the efficiency is affected by the lower oxygen content. Therefore, a flow field that facilitates the distribution of the reagent becomes important. This is demonstrated through the comparison of the performance with the interdigitated flow fields (particularly the A2 type) with those of the parallel and serpentine flow fields.

Independently of the pressure drop, the increase in efficiency in the interdigitated pattern is directly related to the convective effect on the transport mechanism, which increases the amount of reagent that reaches the triple phase region where the reaction takes place. At the same time, the convective flow helps to remove any liquid water formed in the porous electrode structure, whose presence forces the gas to a two-phase flow transport mechanism. These beneficial effects are present to a much lesser extent in the parallel pattern, where the transport mechanism is diffusive, and in the serpentine, where diffusion predominates.

Figure 6 shows the cell voltage-current density curves obtained in single cells operating with hydrogen/air and

with electrode 8 (Table 1), at atmospheric pressure with an air flow corresponding to two times the stoichiometric ($\lambda = 2$) at 2 A cm^{-2}. It is obvious that under those conditions the interdigitated flow field shows the best performance, while the parallel field shows the worst. For $\lambda = 2$ there is not much difference in behaviour among the interdigitated patterns described in Table 2, but among these flow fields the performance seems to be slightly proportional to the contact area gas/electrode.

Figure 7 shows the results for single cells operating under the same conditions but doubling the air flow
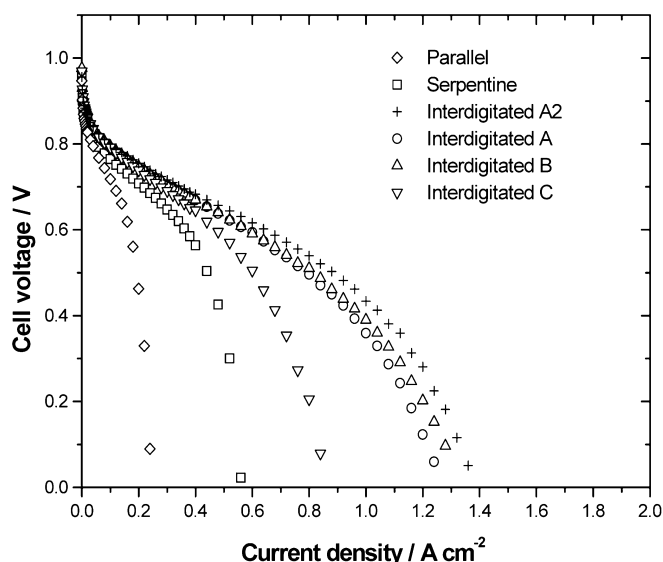

Fig. 6 Cell voltage-current density curves for a single H$_2$/air (at 0.1 MPa) PEMFC at 80 °C with different flow fields and electrode 8. $T(\mathrm{H}_2) = 95$ °C, $T(\mathrm{air}) = 85$ °C, flow of hydrogen $\lambda = 2$ at 2 A cm^{-2}, flow of air $\lambda = 2$ at 2 A cm^{-2}; Nafion 115 membrane
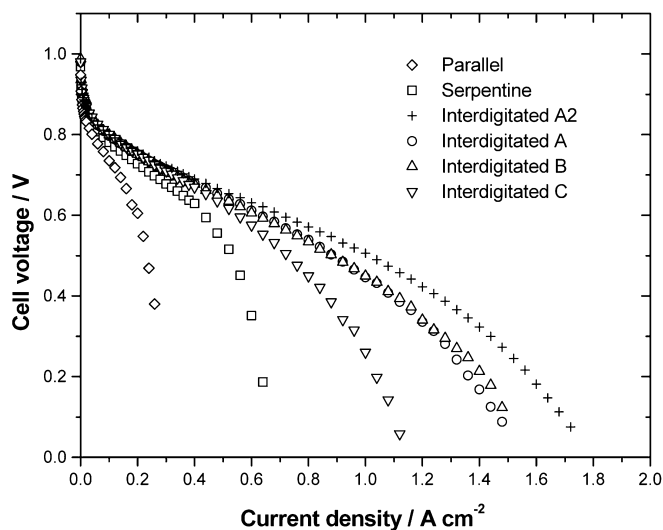

Fig. 7 Cell voltage-current density curves for a single H$_2$/air (at 0.1 MPa) PEMFC at 80 °C with different flow fields and electrode 8. $T(\mathrm{H}_2) = 95$ °C, $T(\mathrm{air}) = 85$ °C, flow of hydrogen $\lambda = 2$ at 2 A cm^{-2}, flow of air $\lambda = 4$ at 2 A cm^{-2}; Nafion 115 membrane

Table 4 Pressure drop in the PEMFC at 80 °C, $T(\mathrm{H}_2) = 95$ °C, $T(\mathrm{O}_2/\mathrm{air}) = 85$ °C, Nafion 115 membrane and different flow fields under the investigated conditions

Flow field	Pressure: 100 kPa[a]			
	$\Delta P(\mathrm{H}_2)$	$\Delta P(\mathrm{O}_2)$	$\Delta P(\mathrm{air})$	
	$\lambda = 2$	$\lambda = 3$	$\lambda = 2$	$\lambda = 4$
Parallel	0	0	0	3
Serpentine	0	0	3	7
Interdigitated A2	3	5	8	21
Interdigitated A	4	6	15	34
Interdigitated B	17	21	38	57
Interdigitated C	18	26	45	72

[a]Absolute pressure at the cell outlet

($\lambda = 4$). The performance of the cell with a parallel flow field is practically the same as that of the cell with $\lambda = 2$, while that of the cell with a serpentine flow field shows a small increase. On the other hand, the effect on cells with interdigitated flow fields is much more pronounced. Now the performance of the cell with type A channels (Table 2) is clearly the best. These are the channels with the smallest dimensions and the ones that should promote a better contact between the reactant gas and the electrode material. It is obvious that under any circumstances an increased air flow should promote the elimination of product water, particularly liquid water. However, if this were the main effect of the increased air

flow it would be expected to be more noticeable in the field with the larger channels. It should be pointed out here that the increase in performance with increasing air flow observed in cells with the interdigitated flow fields is quite dramatic, and in some cases the performance may be two times that shown by a cell with a serpentine flow field. Considering that this latter configuration is used often, the results obtained here concerning the designs of the flow fields may be very relevant when the objective is to improve the performance of the PEMFC.

These observations are summarized in Table 5, where the values of the cell voltage at the maximum power of the cell, and the corresponding current densities, are given for electrode 8 and the two air flows ($\lambda = 2$ and $\lambda = 4$) for the different flow fields.

Figure 8 shows comparative results for the serpentine and the type A2 interdigitated flow fields obtained with electrode 9 (Table 1). Apart from the two air flow conditions ($\lambda = 2$ and $\lambda = 4$), experiments in which the air pressure was increased from atmospheric to 0.3 MPa are also included. Because of the lower performance, the effect of an increased air pressure is much larger in cells with the serpentine flow fields. This can also be observed in Table 6, which shows the values of the cell voltage at the maximum power density, and the corresponding current densities, for the different conditions. On the other hand, it must be noted that the effect of increasing the air flow is small on cells with the serpentine type flow field, at both air pressures, while the effect is still there for cells with the interdigitated flow fields. Although these experiments with air under pressure help in the understanding of the behaviour of cells with different flow fields, it must be pointed out that from the technological point of view working under pressure is undesirable for PEMFCs. Figures 6 and 7 show the advantages of the interdigitated flow field when working at atmospheric pressure. The performance of a cell with an interdigitated flow field working at atmospheric pressure is similar to that of a cell with a serpentine flow field working under a pressure three times higher (Fig. 8), which is an important issue when considering operational fuel cells. It can be demonstrated that the PEMFC pressurization involves an energy cost that hardly compensates the gain in performance.

Table 5 Cell voltage at the maximum power densities and corresponding current densities for electrode 8 for a H_2/air PEMFC with different air flows

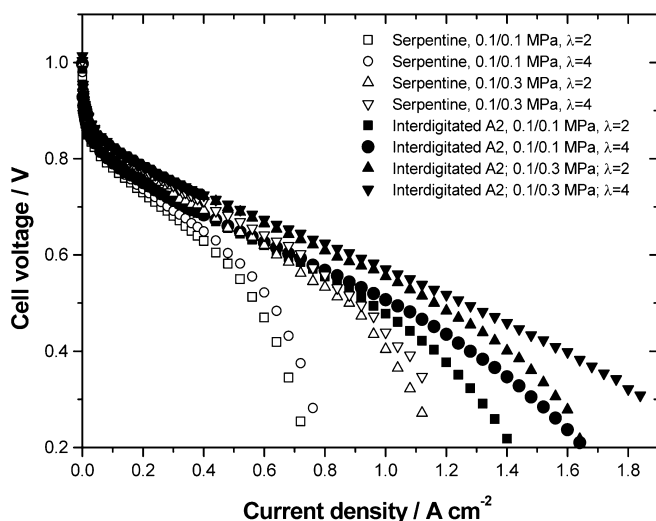| Flow field design	$\lambda = 2$		$\lambda = 4$	
	V	A cm^{-2}	V	A cm^{-2}
Parallel	0.561	0.18	0.548	0.22
Serpentine	0.564	0.40	0.516	0.52
Interdigitated A2	0.462	0.96	0.442	1.16
Interdigitated A	0.450	0.88	0.432	1.04
Interdigitated B	0.487	0.84	0.434	1.04
Interdigitated C	0.505	0.60	0.505	0.72

Fig. 8 Cell voltage-current density curves for a single H_2/air PEMFC at 80 °C with different flow fields, electrode 9 and with different air flows and pressures. $T(H_2) = 95$ °C, $T(air) = 85$ °C; Nafion 115 membrane

Conclusions

The interdigitated A2 design was the flow field that comparatively presented the better performance, for all

Table 6 Cell voltage at the maximum power densities and corresponding current densities for electrode 9 for a H_2/air PEMFC

Flow field design	$\lambda = 2$; 0.1/0.1 MPa		$\lambda = 4$; 0.1/0.1 MPa		$\lambda = 2$; 0.1/0.3 MPa		$\lambda = 4$; 0.1/0.3 MPa	
	V	A cm^{-2}	V	A cm^{-2}	V	A cm^{-2}	V	A cm^{-2}
Serpentine	0.513	0.56	0.522	0.60	0.500	0.88	0.495	0.92
Interdigitated A2	0.461	1.04	0.451	1.16	0.455	1.28	0.446	1.44

the electrode structures and compositions. In some cases, PEMFCs with this flow field presented twice the performance reached in cells with serpentine flow fields. The improvement can be attributed to the combined effects of a convective transport mechanism for the reactant gas together with a better distribution of the channels over the electrode area.

It is also concluded from the results of this work that the better performance is not obtained with just any interdigitated flow field. If the number of channels and their dimensions are not chosen properly, the results may be worse than those of the more common serpentine flow field.

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