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Effect of anode and cathode flow field design on the performance of a direct methanol fuel cell

V.B. Oliveira[a], C.M. Rangel[b], A.M.F.R. Pinto[a,*]

[a] Centro de Estudos de Fenómenos de Transporte, Departamento de Eng. Química, Faculdade de Engenharia da Universidade do Porto, Porto, Portugal
[b] Instituto Nacional de Energia e Geologia, Fuel Cells and Hydrogen, Lisboa, Portugal

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ABSTRACT

In the present work the effect of three different anode and cathode flow field designs (single serpentine (SFF), multi-serpentine (MSFF) and an original design mixed parallel and serpentine (MFF)) on the performance of a DMFC, was investigated experimentally. The studies were conducted in an "in-house" developed DMFC with 25 cm^2 of active membrane area, working near ambient pressure, using two values of methanol concentration (0.75 M and 2 M), of fuel cell temperature (20 °C and 60 °C), of methanol flow rate and of air flow rate. With respect to the anode flow field design it was found that for the two values of methanol flow rate tested, the lower value of fuel cell temperature and lower value of methanol concentration, the use of MFF has a positive effect on cell voltage and power. For the cathode flow field design, for the two values of air flow rate tested, for the lower value of fuel cell temperature and lower value of methanol concentration the use of MSFF leads to a better performance.

For the higher value of methanol concentration tested, a very important condition for portable applications, the use of MSFF or MFF as anode flow field design and MSFF or SFF as cathode flow field design leads to an enhanced fuel cell performance. Most of the reported experiments were conducted close to room temperature, providing information and results that can be used when designing a portable DMFC, where less severe operating conditions are required (ambient temperature and ambient pressure).

1. Introduction

Within the last years, there has been an increased interest in direct polymer electrolyte membrane fuel cells (DPEMFCs). The direct fuel cells (DFCs) use liquid fuels (in liquid or vapor form) directly as a fuel without a reforming step. The most commonly used liquid fuels in direct fuel cells are methanol, ethanol and formic acid. Among different fuel options, methanol is an attractive fuel because it is a liquid at room temperature, has limited toxicity, high energy density, is easy to handle, relatively easy to distribute and has low cost since it can be generated from natural gas, coal, or biomass. Due to its important characteristics, the direct methanol fuel cells (DMFCs) have received in the last years the most extensive attention and efforts compared to other types of DFCs.

The commercialization of the DMFC, is however, still hindered by several technological problems such as slow anode kinetics arising from a multi-step fuel oxidation process at the anode, fuel crossover from anode to cathode. The crossover not only lowers the fuel utilization, but also degrades the cathode performance and generates extra heat. The slow anode kinetics results in higher anodic overpotentials.

While several work continues in order to elucidate the fundamental electrochemical reaction mechanisms, to explore new compositions and structures of catalysts, and to develop new membranes and methods to prevent methanol crossover, important system issues relevant to DMFC are emerging, such as water management, gas management, flow field design, electrode structure and optimization of the operating conditions. The direct methanol fuel cell is a multiphase system involving simultaneous mass, charge and energy transfer. All these processes are intimately coupled, resulting in a need to search for optimal cell design, such as flow field design, and operating conditions, cell temperature, methanol concentration, cathode pressure and methanol and air flow rate. A good understanding of this complex, interacting phenomena is thus essential and can most likely be achieved through a combined mathematical modelling and detailed experimental approach.

To improve the levels of performance in DMFCs, there is an increased interest in reducing mass transport limitations and the kinetic and ohmic limitations. In this regard, some work has been done in order to improve the characteristics of the backing layer in terms of composition and thickness [1–5] and the design of the reactant flow fields [1,6–8]. Once the reactants enter the cell,

* Corresponding author.
E-mail address: apinto@fe.up.pt (A.M.F.R. Pinto).

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Table 1
Comparison of different flow fields used in DMFCs.

Flow field	Advantages	Disadvantages	References
Parallel	Low pressure drops	Inhomogeneous reactant distribution and difficult product removal	[1,7,8]
Spot	Similar to parallel flow field	Similar to parallel flow field and high cell resistance due to a reduced contact to the MEA	[1]
Serpentine	Helpful to remove reaction products and to enhance two-phase mass transport	High pressure drops between the inlet and outlet	[7,8]

they must be distributed over the entire active area. This is typically accomplished through a flow field, which may be in a form of channels covering the entire area in some pattern or porous structures. The main task of these flow fields is to guarantee distribution of fuel and oxidant over the reaction surface area as well as removal of products from the cell. In particular at the DMFC anode, the flow field supplies the methanol solution and transports out CO_2 gas bubbles from the cell. Since the two-phase flow behaviour in the flow field affects the mass transport of methanol to the catalyst layer and the bubbles removal, it has a direct impact on cell performance [9–11]. Different types of flow fields for DMFCs have been presented in the literature [1,6–8]. The comparison between flow fields has been summarized by Qian et al. [12]. The most common designs are spot (or grid), parallel, and serpentine flow channels. Most of the previous studies have been focused on the parallel flow field [11,13–16]. Some authors studied the two-phase flow pattern in parallel channels. They observed that the amount of gas increased rapidly with the current density and accordingly, the flow pattern moved from bubbly to slug and annular regimes causing gas clogging in the channels. Lu et al. [14,15] further investigated the mechanism of bubble dynamics and found that the bubbles were entrapped on the carbon paper by surface tension, growing into larger slugs before detachment to the channels.

The parallel and the spot flow field design generally give comparable performance, though at higher current densities a higher cell voltage at the same current densities was verified when the cell uses the parallel design [1]. This may be partly a consequence of an increased cell resistance due to a reduced contact to the MEA in the spot design. The results also suggest that the mass transfer limiting current for methanol oxidation was reached earlier with the spot design. Although the use of this design shows good results there are certain practical limitations: the fabrication is more difficult and thus more expensive, in operation there is a risk of puncturing the MEA which occurs more frequently if the spots are not precisely aligned on the cathode and anode side of the MEA.

The most widely employed flow field in direct methanol fuel cells is the serpentine configuration. In such a configuration, the reactant is constrained to flow in a zigzag way along parallel channels which are machined in a graphite plate in contact with the electrode backing layer.

The DMFC equipped with serpentine flow field shows lower methanol crossover, higher fuel utilization and a slightly large efficiency at low current densities. When comparing the serpentine and parallel flow field the results presented in the literature [7,8] show that a DMFC equipped with the serpentine flow field shows better performance than those with the parallel flow field. The two-phase flow behaviour in the anode serpentine flow field has also been visually studied [2,8]. Similar periodical repetition of bubble formation, growing and coalescence to gas slugs was found. But under the same conditions no gas clogging phenomena was detected in the serpentine design indicating a superior ability for gas removal [8]. Since the serpentine flow field exhibited a better performance than the parallel flow field some authors [8] focused their studies on the effects of the open ratio and channel length of the serpentine flow field on the cell performance and pressure drop. The results indicated that the open ratio and flow channel length had effects on the cell performance and pressure drop.

A comparison of the different flow field designs used in direct methanol fuel cells, in terms of advantages and disadvantages is given in Table 1 based on the results from the literature. After analysing the information in Table 1, the flow field designs selected for this work, were a single serpentine (SFF), a three channels serpentine or multi-serpentine (MSFF) and a mixed serpentine and parallel flow fields (MFF). The last flow field design described, the MFF is an innovative design and is tested for the first time in a DMFC. The main goal to use this last configuration (MFF), is to combine the advantages of the serpentine and parallel flow field designs. There is a lack of information in literature concerning the use of the multi-serpentine design in DMFCs. The effect of these different flow field designs on the cell performance was evaluated and analyzed by measuring the current–voltage characteristics at ambient pressure. The experiments were performed with an "in-house" developed DMFC with 25 cm^2 of active area.

The experimental results presented in this work are very important especially for the validation of CFD (computational fluid dynamics) models. It should also be mentioned that the vast majority of the experiments were conducted close to room temperature, providing information and results that can be used when designing a portable DMFC, where less severe operating conditions are required (ambient temperature and ambient pressure).

2. Experimental details

The fuel cell test station was manufactured by Fideris Incorporated. In one compact unit, the Methanol Test Kit (MTK) station comprises a methanol handling system, an oxidant gas handling system and a linear electronic load. To obtain the highest performance of the fuel cell test equipment, Fideris Incorporated recommends operation of all equipment using the FCPower software. This software package has been written especially for the fuel cell researcher to provide complete control of all aspects of fuel cell testing. The schematic diagram of the experimental set-up is shown in Fig. 1.

The loadbank subsystem acts as a large variable power resistor which is capable of controlling the amount of impedance by selecting either how much current is passed through the loadbank, the voltage across the loadbank or power dissipated by the loadbank. The computer constantly monitors both current and voltage and these parameters are used to calculate and track the amount of power that the loadbank is dissipating at any one time. This experimental system also provides control over the anode and cathode flow rates, cell operating temperature, air pressure and methanol temperature. The anode flow rate is controlled and measured by a magnetic drive gear pump which allows a maximum methanol solution rate of 85 ml/min. Included in the recirculating loop are a heater equipped with over temperature protection and a cooling system which can cool the methanol solution to ambient temperature. The cathode mass flow is controlled and measured by a mass flow controller (MFC) and the gas flow rate can be set to a maximum of 10 L/min.

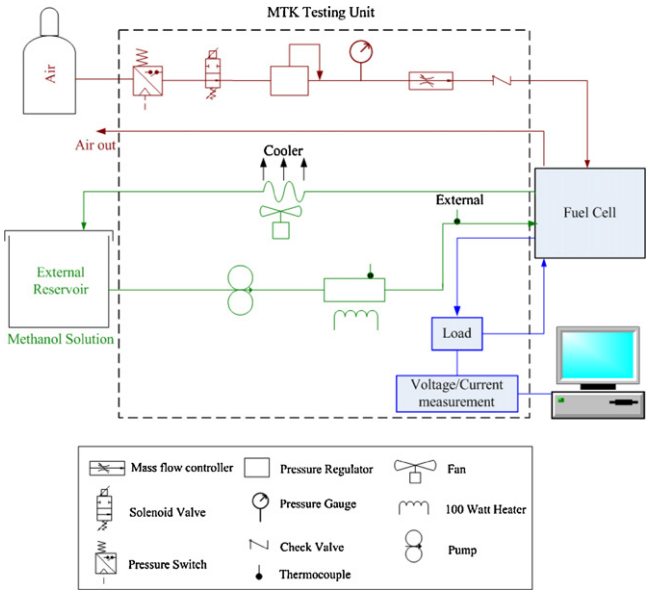

Fig. 1. Schematic diagram of the experimental set-up.

Table 2
Geometry of the different flow fields.

Flow fields	SFF	MSFF	MFF
Channel width (mm)	2.0	2.0	2.0
Channel depth (mm)	2.0	2.0	2.0
Channel length (mm)	50.0	50.0	~20.0
Rib width (mm)	1.5	1.5	1.7
Open ratio (%)	60	60	~60

The experimental fuel cell consists of two aluminium end plates, two gold plated copper connector plates, two monopolar graphite plates with machined flow fields, two diffusion layers, two catalyst layers and a membrane. The membrane used was Nafion 117 (DuPont) with Pt-Ru on the anode side with a loading of 4 mg/cm^2 and Pt-black on the cathode side with a loading of 4 mg/cm^2. The anode and cathode gas diffusion layers used were carbon cloth type A (E-TEK) both with a PTFE content of 30 wt.%. The materials choice follows the conclusions of previous works [18,19].

The three different flow fields are milled into the graphite material HK-3 from Schunk Portugal. The three flow field designs used in this work are presented in Fig. 2. As can be seen in Table 2, the SFF consisted of a single meandering flow channel which has a length of 50 mm and a cross-sectional area of 2 mm × 2 mm, leading to a total cross-sectional area of 4 mm^2. The MSFF consisted of a three meandering flow channel with a cross-sectional area of 2 mm × 2 mm, leading to a total cross-sectional area of 4 mm^2. The MFF consisted of a mixed of parallel and serpentine flow channels with a cross-sectional area of 2 mm × 2 mm, leading, also, to a total cross-sectional area of 4 mm^2. Based on the study from Yang and Zhao [8] care was taken when selecting an adequate open ratio for the configurations used in this work. It should be mentioned that the open ratio of a flow field design is defined as the ratio of the channel area (the exposed portion of the MEA to methanol fuel or oxidant air) to the total MEA area. The three configurations used have an open ratio of 60%, a value similar to the one founded by the authors [8] to be appropriate when the fuel cell is operated under moderate and high methanol flow rates.

3. Results and discussion

The performance of the fuel cell designed in this study was determined by a set of tests, which examined the cell polarization curves and the power density. The tests consisted of applying a load to the cell, measuring the corresponding voltage value and finally calculating the cell power. The influence of the anode and cathode flow field design for different values of methanol concentration, of fuel cell temperature, of methanol flow rate and air flow rate, on the cell performance was carefully investigated for a wide range of operating conditions. A large amount of data was obtained but in order to facilitate the chart reading, a selection of results is here presented for only two values of methanol concentration (0.75 M and 2 M), of fuel cell temperature (20 °C and 60 °C), of methanol flow rate (3 ml/min and 8 ml/min) and air flow rate (1 L/min and 3.6 L/min). The results not presented in this paper showed the same trends and lead to the same conclusions.

Different flow field designs have different resistances due to non-equal contact area between the electrode and the flow field plate [17]. Also different fabrication torques result in different contact resistances. Increasing the fabricating torque gradually generates lowers contact resistance for all the flow field designs. Following the suggestions of Jung et al. [17] the fabrication torque of the cell was maintained at a constant value of 40 kgf cm for all the tested designs. Therefore, the effect on contact resistance due to different flow field combinations on the cell performance can be neglected.

As already referred, the efficient removal of the gaseous carbon dioxide produced by the methanol oxidation at the anode is one of the most important research issues in the development of DMFCs. The carbon dioxide bubbles cause a big pressure loss in the flow fields and also produce gas blocks which keep liquid fuel from diffusing into the catalyst surface. In addition, inhomogeneous

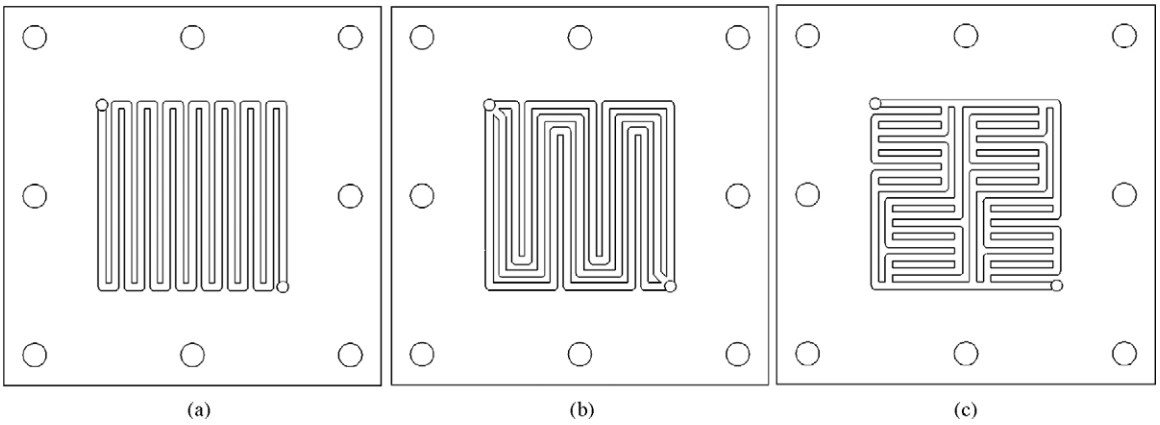

Fig. 2. Designs of the anode and cathode flow fields: (a) serpentine, (b) multi-Serpentine and (c) mixed.

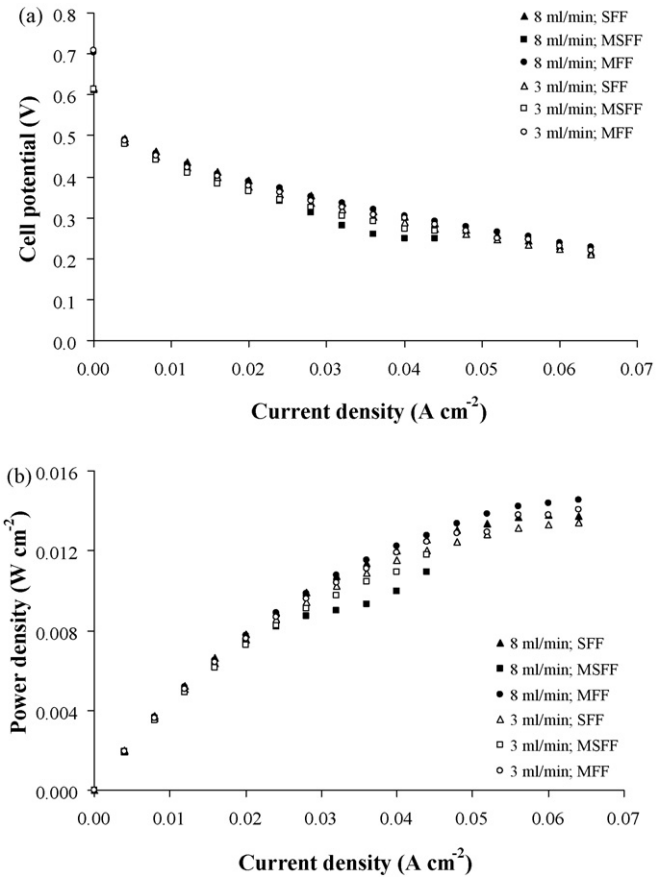

Fig. 3. Effect of anode flow field design on (a) cell performance and (b) power for different anode flow rates. Operating conditions: methanol concentration 0.75 M, air flow rate 3.6 L/min and cell temperature 20 °C.

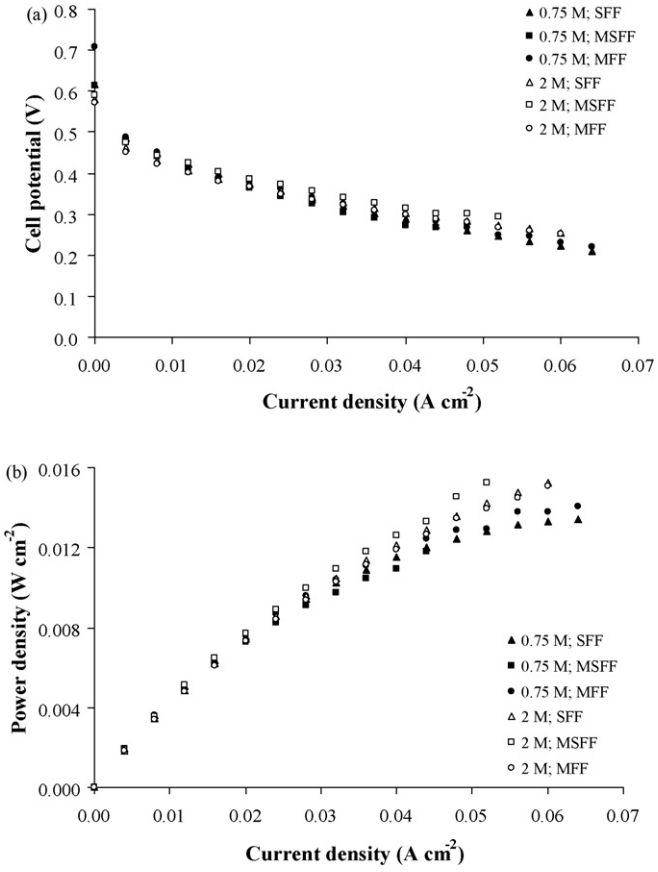

Fig. 4. Effect of anode flow field design on (a) cell performance and (b) power for different methanol concentrations. Operating conditions: air flow rate 3.6 L/min, methanol flow rate 3 ml/min and cell temperature 20 °C.

reactant-gas distribution can occur. The shape of the anode flow field affects the increase of the concentration overpotential due to concentration polarization which is derived from the lack of fuel. The effect of the concentration polarization is stronger at the anode side because of the slow mass transfer rates of the liquid fuel. At the cathode side it is more important to remove water to avoid flooding.

3.1. Anode flow field design

The three anode flow fields were tested by keeping the same cathode flow field having the same configuration and geometry as a SFF.

The data plotted in Fig. 3 were obtained in experiments with a 0.75 M methanol solution fed at two flow rates (3 ml/min and 8 ml/min) and at a fixed fuel cell temperature of 20 °C and air flow rate of 3.6 L/min. The plots show that for the same methanol flow rate the performance of the fuel cell equipped with MFF is slightly better than that with SFF and much better than that with MSFF. The SFF has as the advantage of forcing the reactant flow to traverse the entire active area of the corresponding electrode thereby eliminating areas of stagnant flow. However, this channel layout results in a relatively long reactant flow path, hence a substantial pressure drop and significant concentration gradients from the flow inlet and outlet. Although the MSFF design reduces the reactant pressure drop relative to a single serpentine design, the reactant pressure drop through each of the serpentine channels remains relatively high due to the relatively long flow path of each channel, thus the reactant concentration changes significantly from the flow inlet region to the exit region. Also the MSFF design has more U-bends than the SFF and the CO_2 bubbles tend to be trapped in these regions causing the effective contact area between liquid fuel and the gas diffusion layer to become smaller.

The MFF design is divided in several sections with separate inlet and outlet, and each of flow sectors has parallel flow channels, which are further sub-divided into few sets of channels connected in series. This design gives combined advantages from grid, parallel and serpentine designs and generates lower pressure drops preventing the formation of stagnant flow areas exhibiting distribution of reactants more uniformly with higher average reactant concentrations. These advantages of the MFF result in slightly higher cell performances when operating at high current densities generating a large amount of carbon dioxide bubbles. The bubbles formation raises the pressure drop, so it is advantageous to use a design minimizing pressure drops. The plots from Fig. 3 also show that as expected better performances tend to be obtained for higher methanol flow rates due to a more efficient bubble removal. The exception occurs for the results obtained with the MSFF for the reasons pointed above.

Results from experiments conducted with two different methanol concentrations (0.75 M and 2 M) fed at 3 ml/min, at a fuel cell temperature of 20 °C and air flow rate 3.6 L/min are plotted in Fig. 4. The effect of the methanol concentration on fuel cell performance is discussed in detail elsewhere [18,19]. In general, the impact depends on two opposing effects: increasing concentrations contribute to an increase of the reactant concentration enhancing the anode reactivity and generate high levels of methanol crossover which in turn tend to induce lower performances. For the conditions presented here, the performances at the higher methanol concentration are generally better.

As can be seen from the plots, for a methanol fed concentration of 0.75 M the best performance is achieved with the MFF while for a methanol concentration of 2 M it is achieved with the MSFF. For higher methanol concentrations, the DMFC generates large amounts of methanol crossover which causes a mixed potential on the cathode side and decreases the fuel cell performance. When comparing the three designs, the MFF tends to distribute more uniformly the methanol achieving higher average concentrations all over the flow field area. This homogeneity is an advantage at lower fuel concentrations (0.75 M plot) when the crossover is low. Under high fuel concentrations (2 M plot) the higher fuel concentration achieved induces higher levels of methanol crossover decreasing cell performance. The MSFF design with a less uniform fuel distribution (when compared to the MFF design) and a lower pressure drop when compared to the SFF design) tends to generate lower fuel crossover levels while maintaining a good ability for gas removal and exhibits higher cell performances mainly for higher current densities. It is also useful to analyze the limiting current density for the three cases. The relatively high methanol concentration achieved for the MFF and SFF designs ensures a higher value for the limiting current when compared to the value obtained with the MSFF flow field.

The polarization and power curves represented in Fig. 5 concern experiments with a methanol feed concentration of 0.75 M, a methanol flow rate of 8 ml/min, an air flow rate 3.6 L/min and two different fuel cell temperatures (20 °C and 60 °C). As was already refereed for the DMFC operating at 20 °C, similar performances are achieved with the SFF and the MFF designs with a slight better performance for the latter. An increase on fuel cell temperature leads to an increase of the electrochemical reaction rate. For 60 °C the three anode designs (SFF, MSFF and MFF) give high and similar performances for low current densities. For high current densities the MFF and the SFF have slightly better performance than MSFF. The MSFF design has a relatively long flow path of each channel with a corresponding reactant pressure drop which may cause the appearance of a less uniform distribution of bubbles and in addition presents many U-bend regions. It seems that these characteristics induce a slower gas removal for higher temperatures.

The results presented in this section suggesting the selection of a MFF for the anode flow field for the majority of conditions are summarized in Table 3.

3.2. Cathode flow field design

The three cathode flow fields were tested by keeping the same anode flow field having a SFF configuration.

The design of the cathode flow field affects the air mass transfer rate and the water drainage. If the water is not efficiently removed from the cathode at a sufficient rate, flooding may occur and transport of reactants is hindered [17].

Although at the anode side the advantages of the MFF seem to be superior to the disadvantages, at the cathode side the opposite is found since the performance for all the conditions tested using the MFF is lower. Figs. 6–8 show the polarization and power curves for the three different cathode flow fields.

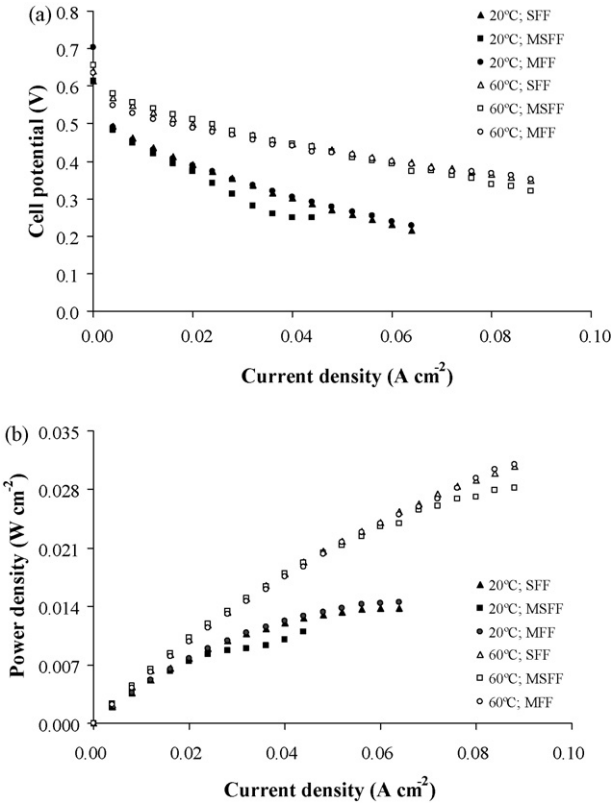

Fig. 5. Effect of anode flow field design on (a) cell performance and (b) power for different fuel cell temperatures. Operating conditions: methanol concentration 0.75 M, methanol flow rate 8 ml/min and air flow rate 3.6 L/min.

Polarization and power curves obtained from experiments with 0.75 M methanol solution fed at 8 ml/min, at a fixed fuel cell temperature of 20 °C and two air flow rates (1 L/min and 3.6 L/min), are presented in Fig. 6. The effect of the air flow rate is as expected: higher performances for higher air flow rates. For the lower air flow rate, both serpentine designs give a good performance due to the presence of the pressure-driven mass flow in the channels ensuring a good ability for water removal. The removal of the water results in the replacement of fresh air which leads to a higher performance depressing the sluggish mass transfer limitations. For the higher air flow rate, the multiple serpentine MSFF exhibits a higher cell performance mainly for higher current densities. Under the high air flow rate used, the water removal is efficient for the two designs but the higher pressure drop in the single serpentine channel is responsible for a more non-homogeneously distributed reactant which is particularly important when operating at low oxygen concentration (air in the present work) and for high current densities (and high reactant consumption). These results are in accordance with the simulation results obtained by Jung et al. [17].

The data plotted in Fig. 7 were obtained in experiments with 0.75 M and 2 M methanol solution fed at 3 ml/min, at a fixed fuel cell

Table 3
Effect of anode flow field design for different operating conditions.

Methanol concentration (M)	Methanol flow rate (ml/min)	Air flow rate (l/min)	Fuel cell temperature (°C)	Best flow field design
0.75	3	3.6	20	MFF
	8			MFF
0.75	3	3.6	20	MFF
2				MSFF
0.75	8	3.6	20	MFF
			60	SSF or MFF

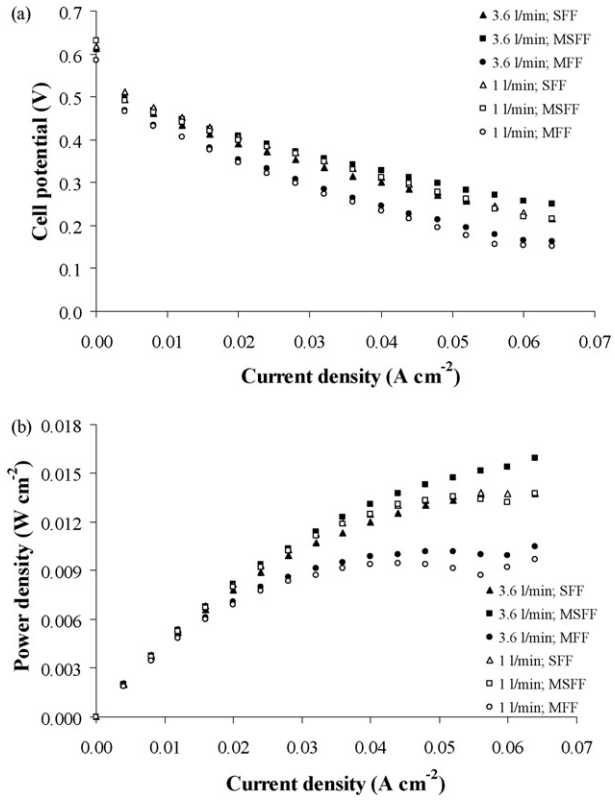

Fig. 6. Effect of cathode flow field design on (a) cell performance and (b) power for different cathode flow rates. Operating conditions: methanol concentration 0.75 M, methanol flow rate 8 ml/min and cell temperature 20 °C.

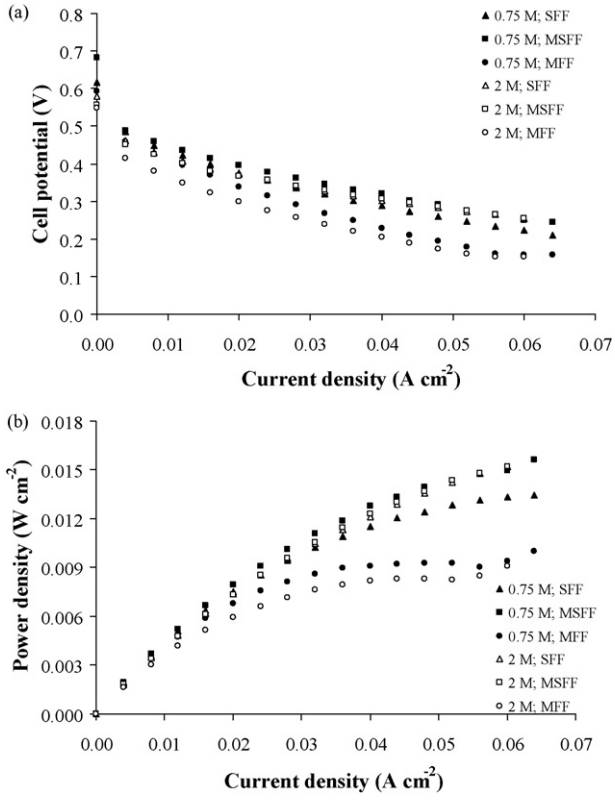

Fig. 7. Effect of cathode flow field design on (a) cell performance and (b) power for different methanol concentrations. Operating conditions: air flow rate 3.6 L/min, methanol flow rate 3 ml/min and cell temperature 20 °C.

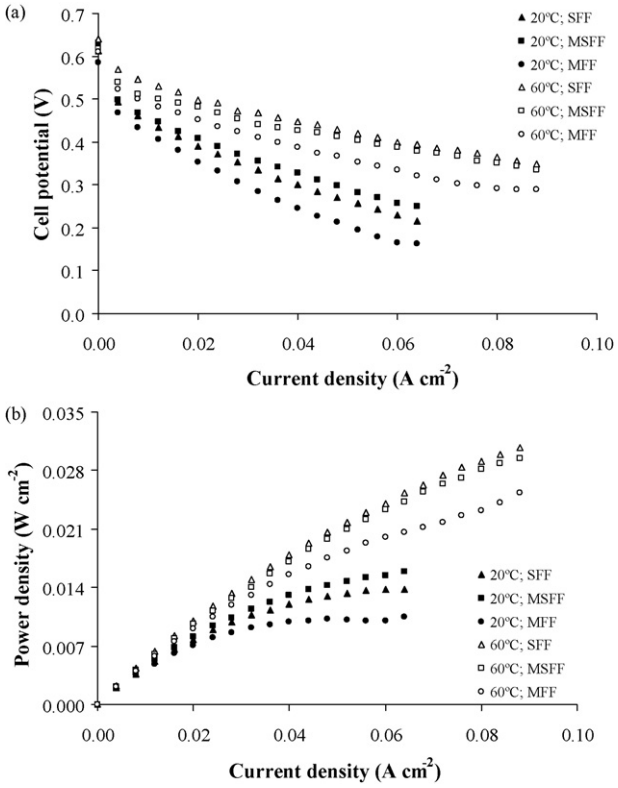

Fig. 8. Effect of cathode flow field design on (a) cell performance and (b) power for different fuel cell temperatures. Operating conditions: methanol concentration 0.75 M, methanol flow rate 8 ml/min and air flow rate 3.6 L/min.

temperature of 20 °C and air flow rates 3.6 L/min. For a methanol fed concentration of 0.75 M the best performance is achieved with MSFF for the reasons explained above while for a methanol concentration of 2 M the higher performances are achieved both with the MSFF and the SFF. For the 2 M methanol solution, the amount of water at the cathode side due to water crossover is smaller. In addition the methanol crossover is higher and oxidizes at the cathode consuming water. With less water present, the advantage of the MSFF over the SFF generating a lower pressure drop and a better distribution of reactant is less important.

For both values of methanol concentration, the MFF design seems to be inefficient in water drops removal at the cathode side, since this design induces much lower pressure drops.

Results from experiments conducted with a methanol feed concentration of 0.75 M, a methanol flow rate of 8 ml/min a air flow rate 3.6 L/min and two different fuel cell temperatures (20 °C and 60 °C), are plotted in Fig. 8. As can be seen from the plots presented in this figure, both MSFF and SFF give enhanced performances when compared to the MFF. As already analyzed, for 20 °C a better performance is achieved using MSFF as cathode flow field design while for 60 °C a slight better performance is achieved using SFF. These results can be explained under the light of the pressure effects on water removal, as explained above. The serpentine design due to the pressure-driven mass flow in the channels allows a correct water removal and forces the reactant flow to traverse the entire active area thereby eliminating areas of stagnant flow. The use of MFF shows the worst performance due to the fact that oxygen and water may flow in one or more of the many channels resulting in a bad distribution of reactant. It should, also, be mentioned that the amount of water in the cathode side is smaller for 60 °C than for 20 °C, since at this temperature more water vaporizes and is removed by the gas stream. In this way a fuel cell operating at 60 °C should have less problems of flooding, so the single serpentine flow

Table 4
Effect of cathode flow field design for different operating conditions.

Methanol concentration (M)	Methanol flow rate (ml/min)	Air flow rate (l/min)	Fuel cell temperature (°C)	Best flow field design
0.75	8	1	20	MSFF
		3.6		MSFF
0.75	3	3.6	20	MSFF
2				MSFF or SFF
0.75	8	3.6	20	MSFF
			60	SSF

fields seems to be more adequate in this conditions, leading to the best performance. Also and as explained before, the smaller amount of water present for the higher temperature makes the MSFF advantage of a more effective oxidant distribution less important.

The results presented in this section suggesting the selection of a MSFF for the cathode flow field for the majority of conditions are summarized in Table 4.

4. Conclusions

The effect of anode and cathode flow field design on the direct methanol fuel cell performance, operating near ambient pressure, has been investigated experimentally. It has been shown that various flow field designs have a large impact on fuel performance and power due to their different ability to provide fuel and remove the produced water and carbon dioxide. From the data reported the following useful information has been obtained:

(i) For the two values of methanol flow rate tested, for low values of fuel cell temperature and low values of methanol concentration the use of an original design mixed parallel and serpentine (MFF) as anode flow field design has a positive effect on cell voltage and power due to the generation of lower pressure drops preventing the formation of stagnant flow areas and exhibiting a more uniform distribution of reactants. For high values of fuel cell temperature the three anode flow field designs used show similar performances.
(ii) For high values of methanol feed concentrations (an important operating condition for portable applications) the use of MSFF as the anode flow field design conducts to a slight better performance than the MFF but generates lower limiting current densities.
(iii) Pressure drop effects at the cathode side are essential to ensure an adequate water removal. In general, both serpentine designs give a good performance at the cathode side ensuring a good ability for removal of water drops. For the two values of air flow rate tested, operating near the ambient temperature and under low values of methanol concentration the use of MSFF as cathode flow field design has a positive effect on cell voltage and power.
(iv) For high values of fuel cell temperature the use of SFF as cathode flow field design conducts to a better performance. For higher values of methanol concentration, an important operating condition for portable applications, the use of SFF and MSFF as cathode flow field design shows similar performances.

In general it was found that for low temperatures and high methanol concentrations, both operating conditions with interest for portable applications the MFF as anode flow field and the MSFF as the cathode flow field lead to enhanced cell performances.

Different flow channel configurations have been proposed in three different designs, including straight channels and serpentine channels. These different flow field designs have advantages and disadvantages which in turn make them suitable for different applications. Improvements in the design of the monopolar or bipolar plates can help achieve the set goals of cost and performance for integration of the DMFC on the market.

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