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Material aspects of the liquid feed direct methanol fuel cell

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A study of a small scale Liquid Feed Direct Methanol Fuel Cell (LFDMFC), based on solid polymer electrolyte membrane, is reported. Two flow cell designs, one with a parallel flow channel arrangement and the other with a spot design of flow bed, are used. The structure of the DMFC comprises a composite of two porous electrocatalytic electrodes; Pt–Ru–carbon catalyst anode and Pt–carbon catalyst cathode, on either side of a solid polymer electrolyte (SPE) membrane. The performance of three Pt–Ru catalysts is compared. The influence of the degree of Teflon loading on the electrode structure is also reported. The effect of the following parameters: cell temperature, oxygen gas or air pressure, methanol liquid flow rate and methanol concentration on the power performance is described.

Keywords: *direct methanol*, *electrocatalysis*, *electrochemical*, *fuel cell*, *platinum ruthenium catalyst*, *polymer electrolyte*

1. Introduction

The direct methanol fuel cell (DMFC) uses methanol, as either vapour or liquid, as fuel and operates at relatively low temperatures ($<100\,^{\circ}\mathrm{C}$). The cell reactions are
anode:

$$CH_3OH + H_2O = CO_2 + 6\,H^+ + 6\,e^- \qquad (1)$$

cathode:

$$\tfrac{3}{2}O_2 + 6\,H^+ + 6\,e^- = 3\,H_2O \qquad (2)$$

The structure of the DMFC (Fig. 1) is a composite of two porous electrocatalytic electrodes on either side of a solid polymer electrolyte (SPE) membrane. The thermodynamic reversible potential for the overall cell reaction is 1.214 V. The direct methanol fuel cell (DMFC) is a promising power source for a range of applications including transportation and portable power sources. It is an alternative to hydrogen fuel cells based on the solid polymer electrolyte which are presently at a significant stage of development particularly for vehicular applications. The DMFC has several features which suit its application to transportation, including good efficiency, low emissions, a potentially renewable fuel source and fast and convenient refueling.

However, a current advantage of the hydrogen cell, over the DMFC, is that hydrogen oxidation at the anode is very fast and, consequently, the performance of the hydrogen cell is better than that of methanol cell. For methanol six electrons must be exchanged for complete oxidation and, consequently, the oxidation kinetics are inherently slower. The slower kinetics are a result of intermediates formed during methanol oxidation [1]. Oxidation of the intermediates to carbon dioxide requires the adsorption of an oxygen containing species (e.g., OH, H_2O). Adsorption of these species does not occur substantially until potentials well above open circuit values [2]. In fuel cells, platinum alone is not a sufficiently active methanol oxidation electrocatalyst and the promotion of methanol oxidation has been actively studied. Currently, significant results have been achieved with the use of binary catalysts, notably Pt–Ru. With these catalysts the second metal forms a surface oxide in the potential range for methanol oxidation [3].

Developments in electrode fabrication techniques and better cell designs for vapour fed cells, have brought dramatic improvements in cell performance in small scale DMFCs. Typically, power densities higher than $0.18\,\mathrm{W\,cm^{-2}}$ are achievable, and power densities higher than $0.3\,\mathrm{W\,cm^{-2}}$ have been reported [4–6]. These power densities are, however, substantially lower than those obtained with H_2 fuel cells, 0.6–$0.7\,\mathrm{W\,cm^{-2}}$, while platinum anode catalyst loading for the hydrogen cells can be substantially lower ($0.1\,\mathrm{mg\,Pt\,cm^{-2}}$).

A limitation in the vapour fed DMFC are the energy requirement to vapourise the aqueous based fuel and the production of carbon dioxide exhaust gas in the fuel. Thus there is a major requirement to separate the unused fuel from the anode exhaust gas containing substantial quantities of carbon dioxide. These factors, together with potential problems in cell thermal management and water management, have focused attention on liquid fed direct, methanol fuel cells (LFDMFC), where carbon dioxide can be simply disengaged from the liquid fuel. In the LFDMFC,

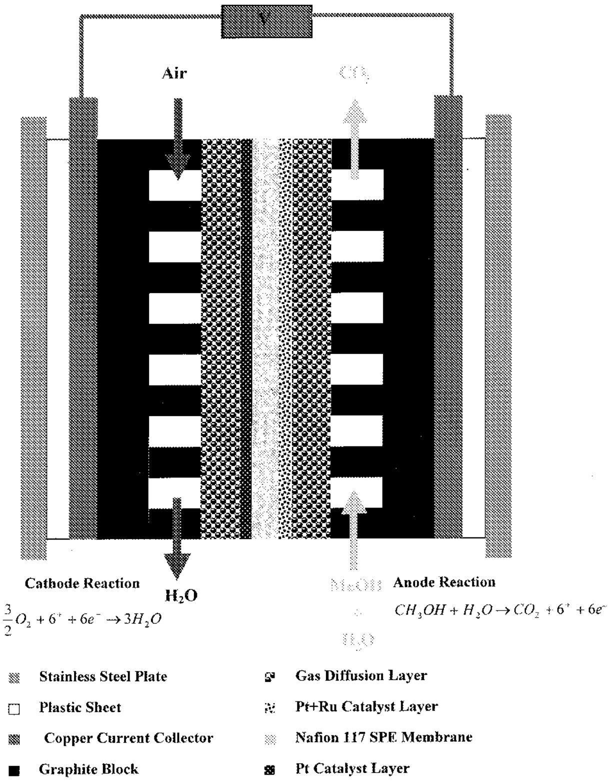

Fig. 1. Schematic diagram of the direct methanol fuel cell.

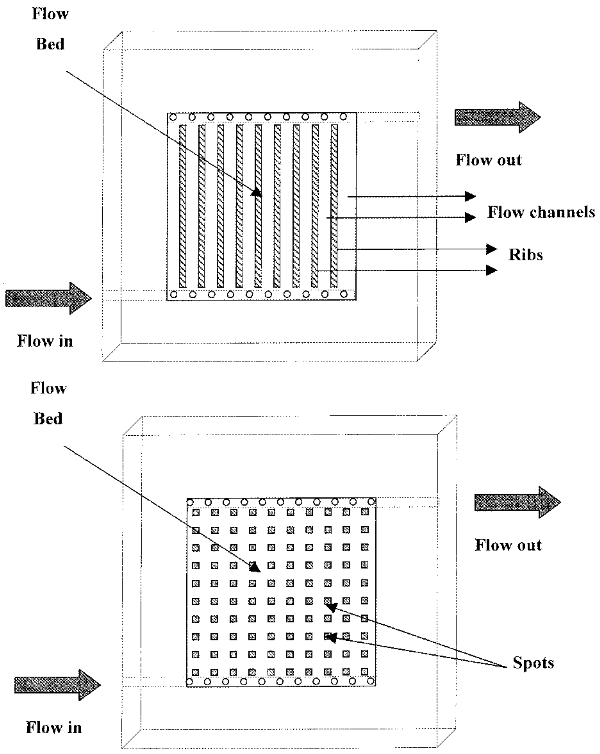

Fig. 2. Schematic diagrams of the DMFC test cell: (a) channel design, (b) spot flow bed design.

methanol is directly oxidised to carbon dioxide using fuel cell electrodes almost identical to those used for vapour fed operation, that is, Nafion®/Teflon® bonded Pt–Ru catalysts dispersed onto carbon. The overvoltages experienced at both electrodes in the DMFC leads to a significant reduction in cell voltage from the theoretical maximum [7–9] and thus research has focused on efforts to minimize these overvoltages. A liquid feed DMFC with power output of 0.2 W cm^{-2} at a temperature of 95 °C and 4 bar oxygen pressure has been reported by Ravikumar and Shukla [10]. Catalyst loading on both electrodes used was 5 mg cm^{-2}.

We have recently reported [11] the performance of a direct methanol fuel cell using aqueous methanol feed with anode catalysts (Pt–Ru) of 2 mg cm^{-2}. In this paper, we report the results of a study of two cell designs, using anode catalyst from three sources. The influence of the Teflon loading of the electrode gas diffusion layer and the presence of acid electrolyte in the methanol feed solution is examined.

2. Experimental details

Tests on the DMFC were performed with two cells, both with a cross–sectional area of 9 cm^2, shown schematically in Fig. 2. The cells were fitted with one membrane electrode assembly (MEA) sandwiched between two graphite blocks with flow paths cut out for methanol and oxygen/air flow. Two types of flow bed were used:

(i) Parallel channel. This consisted of a series of 10 parallel channels, 2 mm deep by 2 mm wide every 1 mm.

(ii) A spot design. This consisted of square spots, 4 mm^2 area, spaced every 1 mm in equally spaced rows and columns. The effective flow channel depth was 2 mm.

The cell was held together between two aluminium backing plates using a set of retaining bolts positioned around the periphery of the cell. Electrical heaters, supplied by Watson Marlow, were placed behind each of the graphite blocks in order to heat the cell to the desired operating temperature. The graphite blocks were also provided with electrical contacts and small holes to accommodate thermocouples. The fuel cells were used in a simple flow rig which consisted of a Watson Marlow perilstatic pump to supply aqueous methanol solution, from a reservoir, to a Eurotherm temperature controller to heat the methanol. Oxygen and air was supplied from cylinders at ambient temperature, and the pressure regulated at inlet by pressure regulating valves. All connections between the cells and equipment were with PTFE tubing, fittings and valves.

MEAs studied in this work were made in the following manner. The anode consisted of a Teflonised carbon cloth support (E–Tek, type A), of 0.35 mm thickness, upon which was spread a thin layer of uncatalysed (ketjenblack 600) carbon, bound with 10 wt % Nafion® from a solution of 5 wt % Nafion® dissolved in a mixture of water and lower aliphatic alcohol's (Aldrich). The catalysed layer, Pt–Ru dispersed on carbon (2 mg cm^{-2} metal loading) and bound with 10 wt % Nafion®, was spread on this diffusion backing layer. A thin layer of Nafion® solution was spread onto the surface of each electrode.

The MEA was obtained by hot pressing the anode and cathode on either side of the pretreated membrane. The thickness of the MEA is approximately 0.8 mm depending on the diffusion layer thickness.

The Pt–Ru anode catalyst used were: (a) Electrochem. Inc (USA); Pt, 20 wt %, Ru 10 wt % on Vulcan XC–72R carbon; (b) Johnson Matthey, Technology Centre development material (UK); 35 wt % Pt, 15 wt % Ru; and (c) In–house catalyst; 40 wt % Pt, 20 wt % Ru on ketjen black carbon. Details of the latter catalyst preparation are described elsewhere [8].

The cathode was constructed using the same method as for the anode, a diffusion layer bound with 10 wt % PTFE and a catalyst layer consisting of 10 wt % Pt on carbon catalyst with a loading 1 mg cm^{-2} Pt black (Johnson Matthey) with 10 wt % Nafion®. The electrodes were placed either side of a pre–treated Nafion® 117 membrane (Aldrich). This pretreatment involved boiling the membrane for 1 h in 5 vol % H_2O_2 and 1 h in 1 M H_2SO_4 before washing in boiling Millipore water (> 18 mΩ) for 2 h with regular changes of water. The assembly was hot pressed at 100 kg cm^{-2} for 3 min at 135 °C. The resulting MEA was installed in the cell after pressing, and hydrated with water circulated over the anode at 75 °C for 48 h. The majority of data reported here were obtained after allowing 48 h to condition a new MEA in the test fuel cell at 75 °C and atmospheric pressure with continuous feed of a 2 mol methanol solution. Several MEAs were tested to ascertain reproducibility of the data. The extent of the conditioning period is of significance in the performance of the cells.

3. Cell performance

Many parameters and variables affect the cell voltage, current density response of the DMFC include; temperature of fuel, methanol concentration, oxygen partial pressure, fuel and oxidant flows, the type of proton exchange membrane, catalyst preparation and the electrode structure and the cell design. In the small cells used in this research, oxidant flows were significantly above stoichiometric requirements and did not influence performance. At low air flow-rates flooding of the cathode structure may possibly occur, restricting access of oxygen to the cathode and thus causing mass transport limitations. These factor will be more important for larger scale operation which is the focus of ongoing research.

Data are reported using the following standard conditions unless otherwise stated:

Methanol concentration: 2 mol dm^{-3}
Methanol solution flow rate: 0.84 cm^3 min^{-1}
Cell temperature: 90 °C
Flow bed: parallel channel
Teflon loading: 13%
Anode catalyst: Electrochem. Inc. Pt, 20 wt %, Ru 10 wt% on Vulcan XC-72R carbon
Air pressure: 2 bar.

In all the data reported the values of open circuit potential were significantly lower than the theoretical thermodynamic maximum; broadly in the region of 700 to 850 mV. Application of an applied current density of approximately 20 mA cm^{-2} resulted in a further loss of 200 mV potential. Further reduction in cell potential, with increased current density, was a result of increased polarisation of electrodes, internal cell resistance and concentration polarization.

Initial performance of the DMFC was investigated using MEAs without Teflon added to the carbon cloth backing layer and which have not been conditioned using methanol flow. The cell performance was not good (Fig. 3) although it did improve with increase in cathode air pressure. After only 2 h of conditioning in methanol there was a noticeable improvement in performance, and further pretreatment

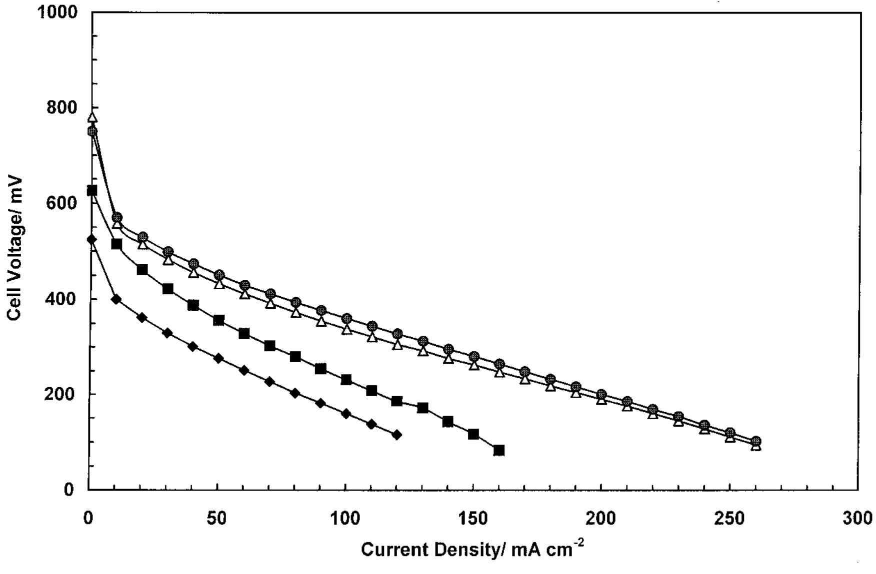

Fig. 3. Effect of 1% Teflon content on the DMFC performance. Key: (△) 2 bar (set 1); (●) 2 bar (set 2); (■) 0.02 bar; (◆) 2.0 bar (unconditioned).

confirmed the requirement for preconditioning of the electrodes for 48 hours. In addition to prescribing a 48 h period of pre-conditioning in methanol solution, the preliminary experimental work also revealed the need for conditioning under current load. From this it is recommended that the electrodes be loaded for 72 h, at 100 mA cm^{-2}, to ensure maximum and reproducible performance. Further improvements in cell performance were expected to be realized by making the carbon cloth hydrophobic, as discussed below.

Figure 3 shows data obtained with a Teflon content of 1% on the carbon cloth. There was a clear improvement in cell performance with the Teflonized backing cloth and the use of a higher air pressure gave a superior performance to that with low pressure (0.02 barg) air.

3.1. *Effect of Teflon loading*

The operation of the DMFC requires that the methanol has good access to the anode while the carbon dioxide gas generated is able to freely move away from the catalyst sites on the catalyst surface. The carbon dioxide gas and aqueous methanol solution move counter currently in the catalyst layer, in the 'gas diffusion' layer and in the carbon cloth backing layer. Ideally these flows should be isolated such that discrete paths for gas flow and for liquid flow exist, rather than a two phase flow with gas bubbles moving against a liquid flow, induced by the anode reaction and the electroosmotic transport of water and methanol. The simplest way to approach this ideal is to make the carbon surface hydrophobic thereby creating regions for free gas movement as routinely adopted in gas fed gas diffusion electrodes. The approach typically is therefore to add Teflon to the carbon cloth, or gas diffusion layers, as adopted in gas fed electrodes. In the case of liquid fed methanol cells the issue of gas flow is more critical than in gas/vapour fed cells due to the 'flooded' nature of the anode. The cathode of the DMFC may be similarly affected by possible problems of flooding, but in comparison to the anode this is a less critical issue.

To explore the effect of anode gas management the Teflon content of the carbon cloth backing layer was varied up to a value of 40% (Fig. 4). At very high Teflon content, the carbon cloth loses conductivity and cell performance is affected by significant electrical resistance losses. The unteflonized cloth produced the poorest performance of all the electrodes. Increasing the Teflon content up to a value of 20% improved cell performance up to current densities of 160 mA cm^{-2}. At higher Teflon content of 30% and 40%, performance of the cell fell. This may be, in part, due to the increased resistance of the Teflonised carbon cloth, as indicated in an increase of the slope of the voltage current density curve with increased Teflon content in the approximate linear, central region of the curve. The Teflon loading which gave the overall best cell performance appears to be around 13 to 20%. The maximum power output of the DMFC with liquid feed was between 90 to 110 mW cm^{-2} using the electrode fabrication and catalyst preparations reported in this paper.

Figure 5 shows data for carbon cloth with a Teflon content of 13% with air at different cell pressures. In the pressure range of 2 bar there was no significant variation in cell performance. Performance was reduced significantly below 1 bar pressure. Also shown in Fig. 5 are data for operation with the unconditioned electrode, which again confirmed the need for electrode conditioning to maximise the cell performance.

The issue of cathode gas operating pressure is clearly an important factor in DMFC operation

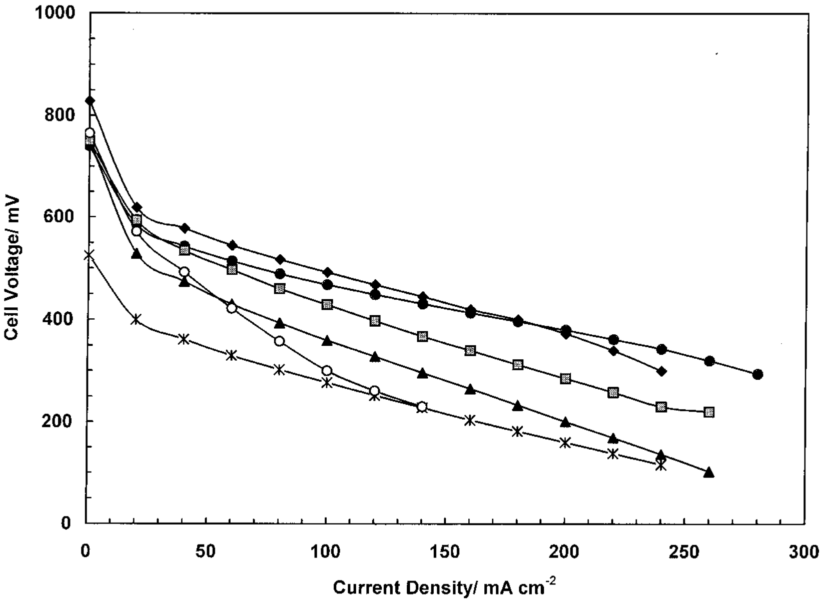

Fig. 4. Influence of Teflon content in the backing layer on DMFC cell voltage characteristics. Teflon content: (∗) 0%, (▲) 1%, (●) 13%, (◆) 20%, (▨) 30% and (○) 40%.

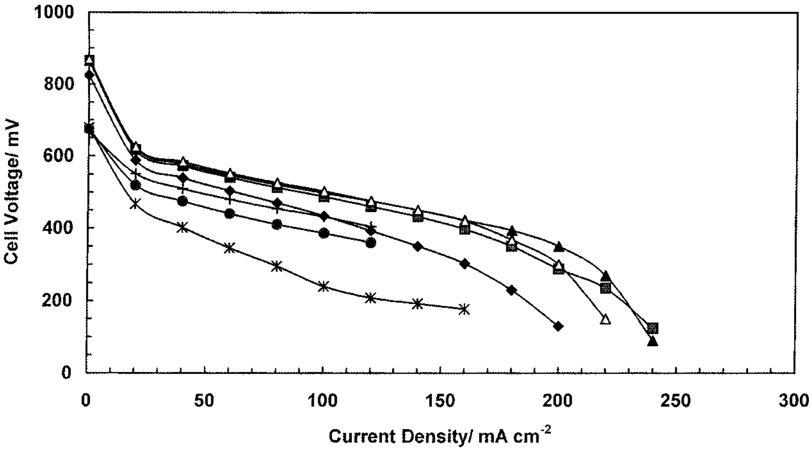

Fig. 5. Effect of air pressure with a 13% Teflonized carbon backing layer. Cathode air pressure (barg): (◆) 0.5, (■) 1, (▲) 1.5, (△) 2, (∗) 0.2 (unconditioned), (●) 0.5 (unconditioned) and (+) 2 bar (unconditioned).

which will be determined from an engineering optimization of the fuel cell power plant, which is outside the scope of this paper. Although it is potentially attractive to utilize pure oxygen to maximise power output, in many applications this is not practical and air fed cathodes are the only option.

Overall at the moment the performance of liquid feed direct methanol fuel cells is not as good as the vapour fed counterparts; maximum power densities are some 50% lower. This is partly due to the restricted operating temperature range used in the liquid fed cells (< 100 °C) and to factors associated with the use of liquids in electrode assemblies originally designed for gas and vapour fed cells.

3.2. *Effect of flow bed design*

A series of tests was performed to investigate the effect of air pressure, methanol fuel flow rate and methanol concentration on cell performance with the spot design of flow bed. The spot design reduces the contact area between the MEA and current collector enabling greater access of fuel to the electrode and greater cross sectional area for carbon dioxide gas release. The spot design may provide improved mixing of the anode fluid at high flow rates. At low flow rates bubble releases from the spot faces normal to the flow may be restricted. However, the reduced contact area to the MEA may make the current distribution over the electrode surface less uniform and increase the internal resistance of the cell.

Figure 6 shows the effect of methanol flow rates in the range 0.84 to 6.96 $\mathrm{cm^3\,min^{-1}}$ on cell performance. These flow rates are relatively low and are equivalent to velocities in the flow channels of approximately 0.028 to 0.3 $\mathrm{cm\,s^{-1}}$ and Reynolds numbers of 0.8 to 7.0 based on the wetted perimeter of the channels. The methanol flow rates used ensured that the residence times in the cell were relatively high, up to 75 s, giving conversions of methanol of up to 12%, for the lowest methanol concentration used (0.5 $\mathrm{mol\,dm^{-3}}$). This, in turn, created large quantities of carbon dioxide gas in the fuel stream and, at high current densities, large gas fractions, up to a theoretical 0.6.

Figure 6(a) shows the results obtained for operation with 2 $\mathrm{mol\,dm^{-3}}$ methanol concentration with flow rates of 0.84 to 2.09 $\mathrm{cm^3\,min^{-1}}$. The lower flow rate gave a slightly better cell performance; the difference in potential at a given current density is approximately 25 mV in the range of flow rates used. At higher current densities the effect of flow rate on cell performance was less significant which may be indicate starvation of methanol at the anode at higher current densities where current started to fall due to a mass transport limitations. Figure 6(b) shows data for the effect of flow rate with a 0.5 $\mathrm{mol\,dm^{-3}}$ methanol solution. The effect of flow rate on cell performance was similar to that with a 2 $\mathrm{mol\,dm^{-3}}$ methanol solution up to a current density of approximately 150 $\mathrm{mA\,cm^{-2}}$, whence there was a rapid fall of cell voltage with current density and the current density approaches a limiting value. Up to this point the performance of the cell with the two methanol concentrations was very similar.

The limiting current density was presumably a combined affect of a lower methanol concentration, coupled with the production of carbon dioxide, in the vicinity of the electrocatalyst. At the lowest methanol concentration used, 0.5 $\mathrm{mol\,dm^{-3}}$, the conversion of methanol approached 10%, and the volume fraction of carbon dioxide was approximately 0.4 at the cell outlet, at the highest current density imposed. At low current densities methanol depletion by mass transport is less significant and the cell benefited from a reduced crossover of methanol from anode to cathode at the lower methanol concentrations. That is, there was a reduced cathode polarization due to a smaller mixed potential. This is seen in the data at a current density of 25 $\mathrm{mA\,cm^{-2}}$ and at open circuit where voltages were some 40 to 50 mV greater at the lower methanol concentration. At the higher value of methanol concentration the mixed potential becomes

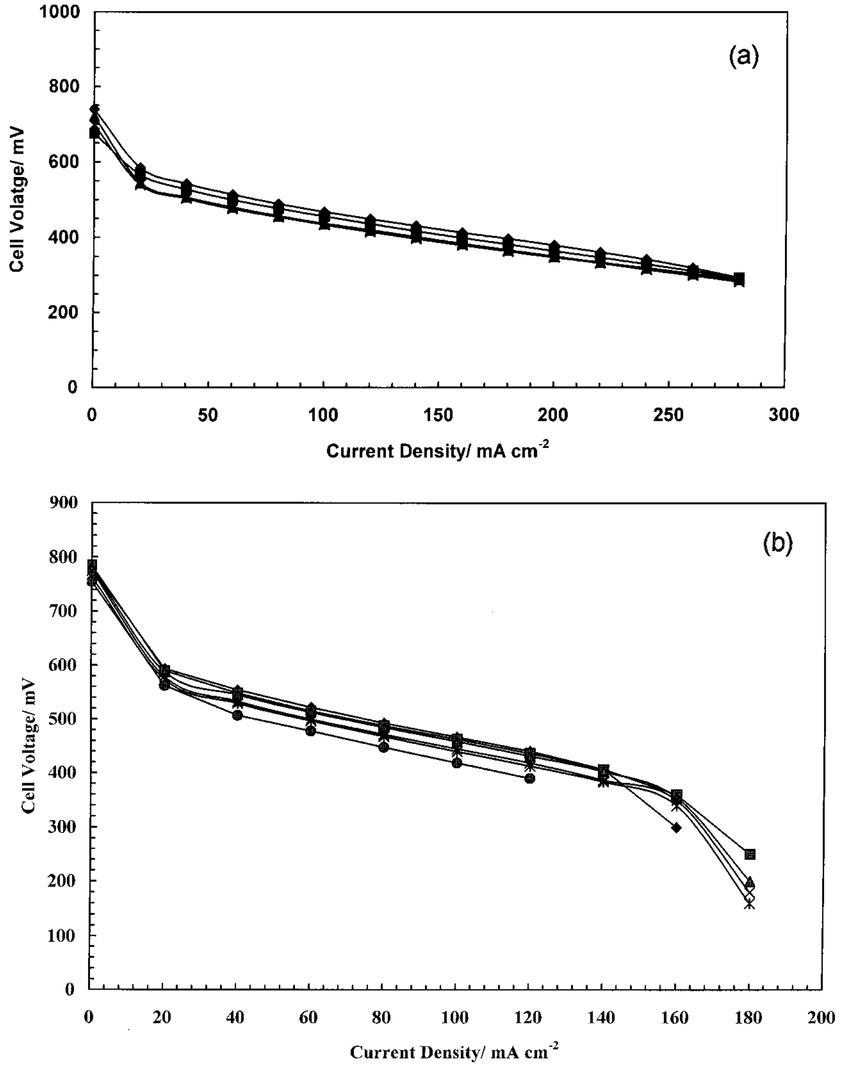

Fig. 6. Effect of methanol flow rate on the voltage, current density response of the DMFC. 90 °C. For (a) 2.0 kmol m^{-3} methanol flow rates are: (◆) 0.8358, (■) 0.6965, (▲) 1.393 and (×) 2.0895 cm^3 min^{-1}. For (b) 0.5 kmol m^{-3} methanol flow rates are: (◆) 0.8358, (▲) 2.0895, (*) 3.4825, (■) 1.393, (×) 2.786 and (●) 6.965 cm^3 min^{-1}.

a more dominant factor over a much wider range of current densities.

Figure 7 compares the performance of the two flow bed designs at different air pressures. The two flow beds generally gave comparable performance, although at the higher current densities there is an indication that the channel flow bed design gave higher cell voltages at the same current densities. This may be partly due to an increased cell resistance due to a reduced contact to the MEA in the spot design. The data also possibly suggests that the mass transfer limiting current for methanol oxidation was reached earlier with the spot design. This may be due to poorer carbon dioxide gas release from the cloth surface, although this would require more detailed evaluation to substantiate this behaviour.

Although the use of a spot flow bed design gave encouraging results, there are certain practical limitations in the design. First, fabrication is more difficult and thus will be more expensive. Secondly, in operation there is a risk of puncturing of the MEA, which occurred on occasions when spots were not precisely aligned on the cathode and anode sides of the MEA. On scale up, this is likely to be a more critical issue which may prevent adoption of the spot design. The parallel flow channel design can be set up with cathode flow channels and anode flow channels at 90° to each other to minimize the risk of MEA puncture.

3.3. *Effect of anode catalyst*

Figure 8 shows cell voltage data for three types of anode catalysts using air fed cathodes. Data for the Newcastle catalyst and Electrochem. catalyst was comparable under the conditions used.

Figure 9 presents data for the effect of cell temperatures (60 and 100 °C) on performance with an air fed cathode for the Electrochem. catalyst. The best performance was achieved at the highest temperature of 90 °C. In the range 60–90 °C there was an approximate 40 mV drop in cell potential per 10 °C fall in temperature. This meant that at 60 °C the maximum power density is approximately 40 mW cm^{-2}.

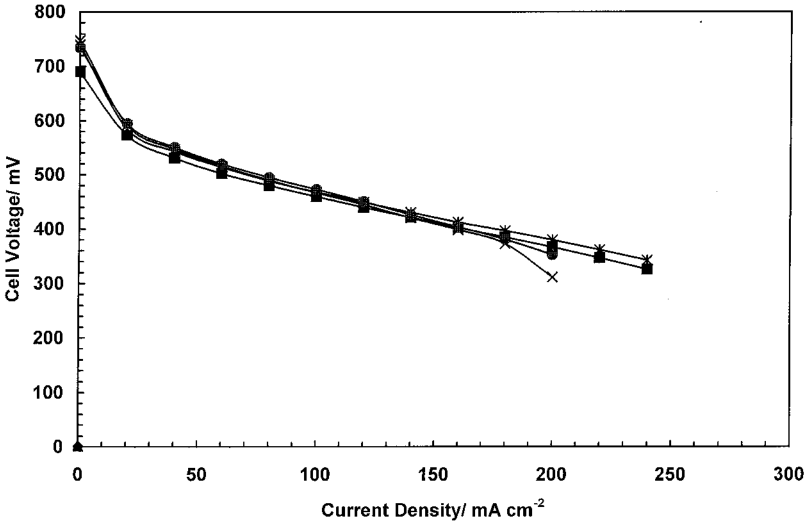

Fig. 7. Comparison of flow bed design on the cell voltage, current density response of the DMFC. Air pressure and bed design: (■) channels 1.5 bar, (×) spots 1.5 bar, (∗) channels 2 bar, (●) spots 2.0 bar.

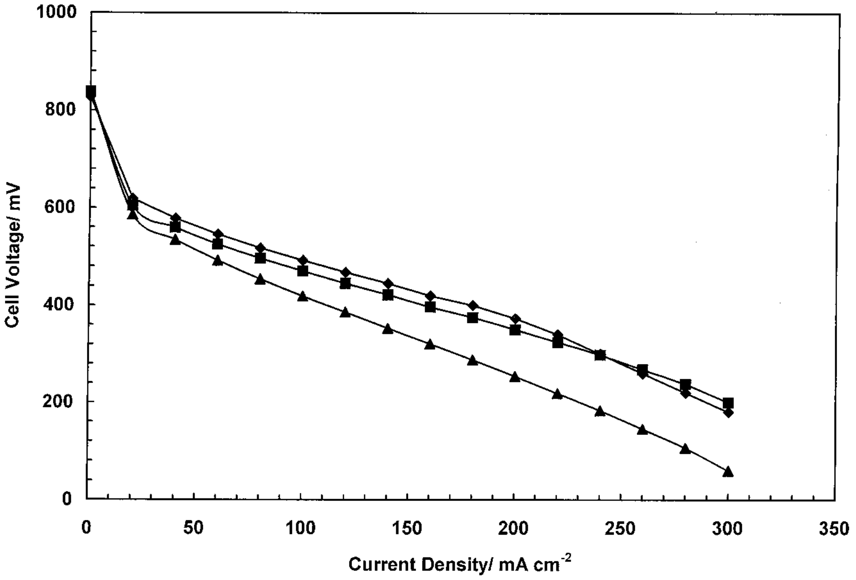

Fig. 8. Effect of anode catalyst on the cell voltage characteristics of the DMFC. Key: (◆) Electrochem; (■) Newcastle; (▲) Johnson Matthey.

3.4. *Influence of acidified methanol*

An attraction of the DMFC, based on solid polymer electrolytes, is that no electrolyte is required in the methanol solution feed. This, in principle, simplifies the operation of the cell system and puts less stringent demands on materials for the stack systems. In the case of a liquid fed cell the addition of electrolyte to the anode feed is now possible and this may influence the performance of the anode catalyst layer. For example, the increase in solution conductivity may improve the current distribution in the anode catalyst layer and, improve the utilisation of the catalyst surface. On the other hand the decrease in pH (higher proton concentration) and the addition of anions may have a detrimental effect on the catalyst performance. Figure 10 shows the effect of phosphoric acid on the cell performance with the Electrochem. catalyst. With the relatively high acid concentrations used, a reduction in cell voltage was seen. This reduction was greater with the higher concentration of acid. After operation with acidified methanol solution, the cell was flushed with aqueous methanol solution and restored to normal operation with aqueous methanol, after which there appeared to be little detrimental effect to the cell. Overall these preliminary data suggests that there is no benefit in the use of relatively high acid concentrations in the methanol solutions with solid polymer electrolyte fuel cells. This data concurs with earlier observations on methanol oxidation in acidified electrolyte where superior performance is achieved in the absence of acid electrolyte with solid polymer electrolyte membranes [12].

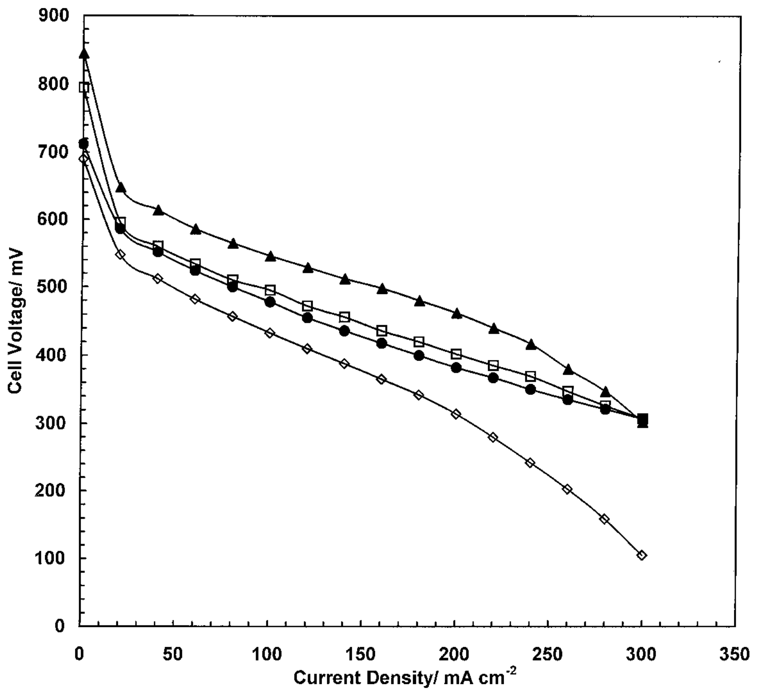

Fig. 9. Effect of temperature on the cell voltage characteristics of the DMFC. Temperatures and cathode gas conditions: (▲) 90 °C, oxygen (□) 90 °C, air (●) 65 °C, oxygen (◇) 65 °C, air.

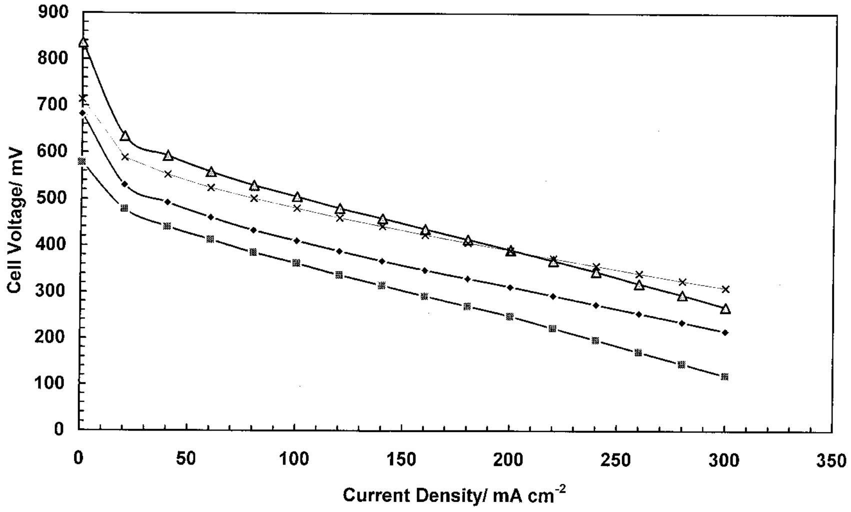

Fig. 10. Effect of phosphoric acid addition to the methanol solution. Acid concentrations (M): (◆) 0.5 M, (■) 2.0 M, (△) no acid, (x) cleaned cell.

4. Conclusions

This research has shown that suitable performance of the LFDMFC is dependent upon materials for the membrane electrode assemblies and on the flow bed design. Variables such as cell temperature and cathode oxygen gas supply have a significant effect on cell performance. Highest power densities are achieved at higher temperatures and cathode gas pressures. The data suggests that an acceptable performance is achieved with operating temperatures of 70 °C and above, but this will depend upon the specific application of the cell. For high power applications, such as in transportation, maximum sustainable operating temperatures are likely to be employed, which may require the use of pressurised anolyte feed to enable operation above temperatures of 100 °C. Overall there is not a considerable effect of liquid flow rate, over the range considered, on the cell performance. Similarly, the influence of methanol concentration is not critical within the range of values studied. A more critical issue is the aspect of methanol conversion in the cell and thus the amount of carbon dioxide produced in the cell.

The performance of the DMFC has been evaluated over several months of operation on an intermittent basis. The ability of the cell electrodes to respond to change in load has been established through a general reproducibility of the performance. Short term continuous tests (72–120 h) showed that the cell performance was stable over a fairly long period of time. The maximum power output of the DMCF with liquid feed was between 90 to 110 mW cm^{-2} using the electrode fabrication and catalyst preparations reported in this paper.

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