

Short communication

Performance of DMFC with SS 316 bipolar/end plates

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Abstract

This work mainly emphasizes the development of new materials and design for a bipolar/end plate in a direct methanol fuel cell (DMFC). According to the DOE requirements, preliminary studies show that SS 316 (Stainless Steel 316) is a suitable candidate. Several flow field designs were studied and a modified serpentine design was proposed. SS 316 end plates were fabricated with an intricate modified serpentine flow field design on it. The performance of a single stack DMFC with SS 316 end plates were studied with different operational parameters. A long-term test was carried out for 100 h with recycling the methanol and the contaminants in the MEA were characterized. The stack efficiency is found to be 51% and polarization losses are discussed. SS 316 with low permeability resulted in an increased pressure drop across the flow field, which increased the fuel cell performance. The use of SS 316 as bipolar plate material will reduce the machining cost as well as volume of the fuel cell stack.

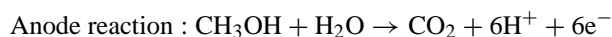
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Keywords: DMFC; SS 316; Bipolar/end plate; Serpentine flow field design

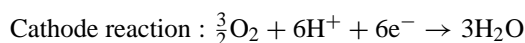
1. Introduction

In recent years, the DOE has focused on fuel cell technology as a means to achieve more efficient and reliable forms of energy conversion. Fuel cells seem to show promise towards achieving the goal. A fuel cell is an electrochemical device, which efficiently converts the chemical energy of fuel directly to electrical energy without combustion. The fuel can be derivatives of higher order hydrocarbons (i.e. H₂, CH₃OH, and CH₄). In 1839, fuel cell was invented by William Grove. Since then, the fuel cell has been attractive due to high energy converting efficiency with nearly zero emission. Fuel cell research is mainly focused as an alternative energy source to a battery or an internal combustion engine. In comparison to a battery, a fuel cell is inexhaustible and works as long as fuel is supplied, whereas a battery is exhaustible and needs to be replaced with time. Several issues for the fuel cell such as component cost, weight, properties, performance at fuel cell environment and fuel storage need to be resolved before the advantages of fuel cell can be exploited.

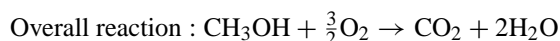
Fuel cells are classified in to different groups based on their components, functions and applications. Among these, DMFC and PEMFC can be used for low temperature applications. DMFC works with liquid methanol unlike PEMFC which uses H₂, and thereby eliminates the on-board H₂ storage problem. As a result, the risk of explosion in the use of fuel cells is eliminated. Liquid methanol is suitable for storage. DMFC can be considered economical, as the fuel (liquid methanol) can be recycled and external humidification can be avoided. In some places, DMFC works without external parasitic losses and works by capillary action, buoyancy and diffusion processes. The electrochemical reactions involved in DMFC are given as follows [1].



$$E_1^\circ = 0.016 \text{ V/SHE}$$



$$E_2^\circ = 1.229 \text{ V/SHE}$$



$$E_{\text{eq}}^\circ = 1.21 \text{ V}$$

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$$\Delta H^0 = -726 \text{ kJ (exothermic reaction)}$$

where SHE refers to standard hydrogen electrode.

The challenges for efficient DMFC performance include: (1) methanol crossover; (2) slow chemical kinetics; (3) carbon dioxide production at anode; (4) poisoning of the catalysts [2,3]. The effect of all these problems can be reduced by improving certain components like the bipolar/end plate material and its design, membrane electrode assembly (MEA) and the catalyst loading.

Bipolar/end plates are the most important and the costliest components in a fuel cell. The component alone constitutes 80–85% of the total weight of the cell stack. It is called an end plate for a single stack and functions as a bipolar plate for a multi-stack fuel cell having flow field design on both sides of the plate. A bipolar plate acts as a separator plate between two cells which keeps methanol and oxygen from direct contact. The intricate flow field design in bipolar/end plate distributes the fuel (liquid methanol and oxygen) uniformly over the reaction surface. The material properties and intricate flow field design of bipolar/end plate plays a vital role in the efficiency of the DMFC. As per the target of DOE, the bipolar plate material for a fuel cell must be low cost ($<10\text{US}\$\text{kW}^{-1}$), low weight ($<1 \text{ kg kW}^{-1}$ of system), corrosion resistant ($<16 \mu\text{A cm}^{-2}$), highly conductive ($>100 \text{ S cm}^{-1}$) and impermeable ($<2 \times 10^{-6} \text{ cm}^3 \text{ cm}^{-2} \text{ s}$) [4]. A perfect flow field design should distribute liquid methanol and oxygen gas uniformly to the anode and cathode reaction surfaces, respectively. The electrochemical reaction that occurs in a DMFC is exothermic and produces heat during the reaction. The design should efficiently remove the heat produced by the chemical reaction, avoiding local build up of heat. The anode reaction in the DMFC has a byproduct, CO_2 , which inhibits the motion of methanol in the flow field. This leads to a significant reduction of active area which reduces the overall performance of the fuel cell stack. The efficiency of flow field design can also be influenced by the permeability of fuel in bipolar/end plate material. Material with low permeability for fuel gives a better performance in fuel cell environment as this increases the pressure drop across the flow field. The increase in pressure drop at the contact between the fuel and interface changes the diffusion process to a convective process at the interface [5,6]. As a result, the fuel cell gives better performance.

Graphite is currently used as the commercial bipolar/end plate material. It has poor mechanical properties, which increase the machining cost and also limits the plate thickness. To meet the stiff challenge of cost, weight and volume issues with bipolar/end plates, our research is focused on finding metals with low weight, cost and which can be easily machined in to thin plates. Metallic components have higher conductivity, better mechanical properties and thus can be easily machined at less cost, with smaller volume relative to the graphite.

2. Experimental

2.1. Bipolar/end plate material and design

Several materials were investigated, which could be possible alternatives to commercial graphite as a bipolar/end plate material. Concentrating on different factors like cost, density, mechanical strength and availability, SS 316 (Stainless Steel 316) was chosen as the alternative. Table 1 shows the comparison between properties of graphite and SS 316 as a material for the bipolar/end plate [7]. It indicates that SS 316 has superior properties as compared to commercially used graphite. However, SS 316 shows weak corrosion resistance unlike graphite which can be avoided by surface modification methods [8,9], which is an ongoing research in our laboratory.

Several flow field designs like serpentine parallel, and interdigitated have been studied. A serpentine design was chosen for its superior performance in the DMFC environment [10,11]. In this design, the production of CO_2 at the anode does not reduce the active area unlike the parallel design. A modified serpentine design was developed for our experiments from the serpentine design. The modified serpentine design has multi-parallel grooves in a serpentine pattern as shown in Fig. 1.

2.2. Bipolar/end plate fabrication and fuel cell stack integration

A single stack DMFC was developed at our laboratory. Since the experiments were conducted on a single stack DMFC, end plates were fabricated instead of bipolar plates. The end plates made of SS 316 were machined with an intricate modified serpentine flow field design as shown in Fig. 1. The active area of the flow field was 25 cm^2 .

The single stack of DMFC includes two aluminum-supporting plates for clamping the end plates as shown in Fig. 2. A current collector of Al 6061 was joined with SS 316 end plates. The MEA was purchased from BCS Technologies with 2 mg cm^{-2} Pt-Ru catalytic loading on the anode and 0.4 mg cm^{-2} Pt loading on the cathode and NAFION-117 as membrane. The MEA was sandwiched between the two end plates. The single stack DMFC along with the an-

Table 1
Comparison of properties between Graphite and SS 316 [7]

| Property | Graphite | SS-316 |
|---|-------------------------------|-------------|
| Cost ($\text{\$ kg}^{-1}$) | 75 | 7 |
| Density (g cc^{-1}) | 2.25 | 8.02 |
| Thickness of bipolar plate for same weight (mm) | 2.5–4 | 1 |
| Modulus of elasticity (GPa) | 10 | 193 |
| Tensile strength (MPa) | 15.85 | 515 |
| Corrosion current (mA cm^{-2}) | <0.01 | <0.1 |
| Electrical resistivity ($\Omega \text{ cm} \times 10^{-6}$) | 6000 | 73 |
| Thermal conductivity (W (m K)^{-1}) | 23.9 | 16.3 |
| Permeability ($\text{cm}^3 \text{ cm}^{-2} \text{ s}$) | Porous 10^{-2} to 10^{-6} | $<10^{-12}$ |

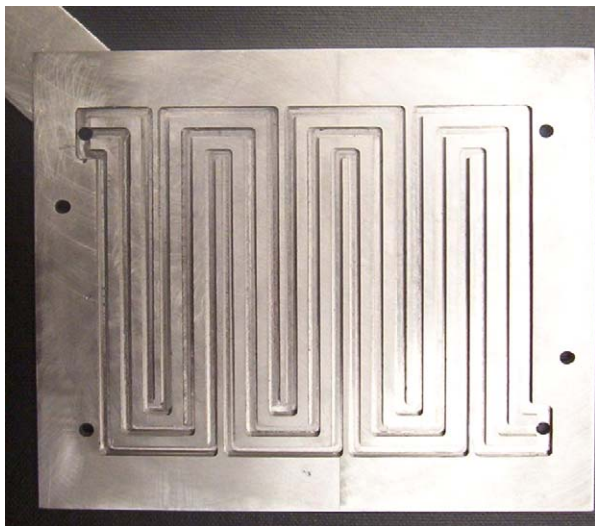


Fig. 1. Fabricated SS 316 end plate with modified serpentine flow field design.

ode and cathode fuels were preheated to 70 °C by inserting a heating coil in the aluminum supporting plates.

All the experiments were carried out by using the fuel cell station. The fuel cell station includes: (1) CompuCell GT[®] gas controller unit for precise control of mass flow of oxygen as well as humidification, temperature and back pressure of oxidant; (2) Scribner Associates 890B fuel cell load system for precisely drawing desired amount of current from the stack; (3) micro fuel pump to pump and heat the liquid methanol as per requirement; (4) FuelCell[™] software for precise computer control and monitoring the operating parameters.

2.3. Experimental procedure

Experiments were carried out to check the performance of a single stack DMFC with SS 316 end plates. The DMFC stack was tested for different operational parameters like temperature and pressure. The experiments include: (1) a long-term test for open circuit voltage (OCV); (2) polarization studies; (3) effect of temperature; (4) effect of pressure on single stack DMFC. These were observed by optimizing the concentration of methanol that 2 M methanol gave better performance and led to lesser methanol crossover compared to other concentrations [10]. 2 M methanol were used as the fuel for experiments. The methanol was recycled throughout the experiments. Pure oxygen with 99.8% purity was used as the reactant gas at the cathode. The experiments were carried out by keeping the anode at atmospheric pressure and the fuel flow rate was kept constant at the anode and cathode at 100 cc min⁻¹ and 70 cc min⁻¹ + LBF (load based flow), respectively.

3. Results and discussion

3.1. Long-term test

A long-term OCV test was carried out for 100 h and a consistent output was obtained without any significant drop in voltage. The test was done to check the output without any load. The operating conditions were $T = 70\text{ }^{\circ}\text{C}$; $P = 200\text{ kPa}$ (at the cathode side). An initial voltage of 0.795 V was obtained and it decreased to a consistent open circuit voltage (OCV) of 0.614 V along with time. The performance of the

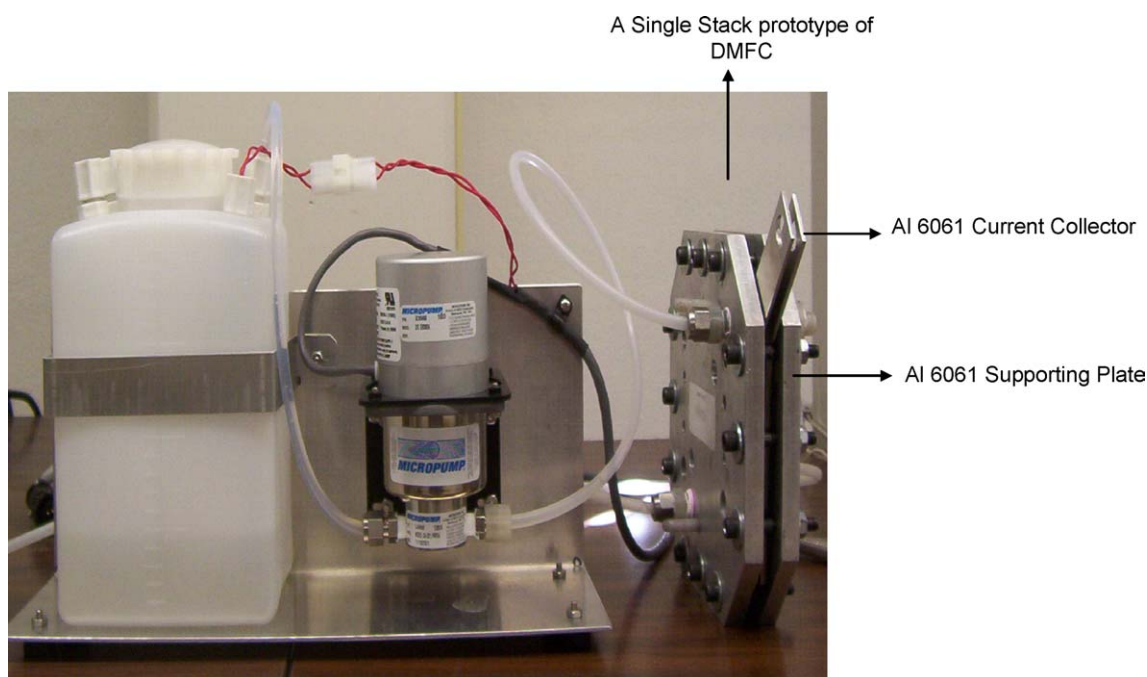


Fig. 2. A single stack prototype of DMFC developed at our laboratory.

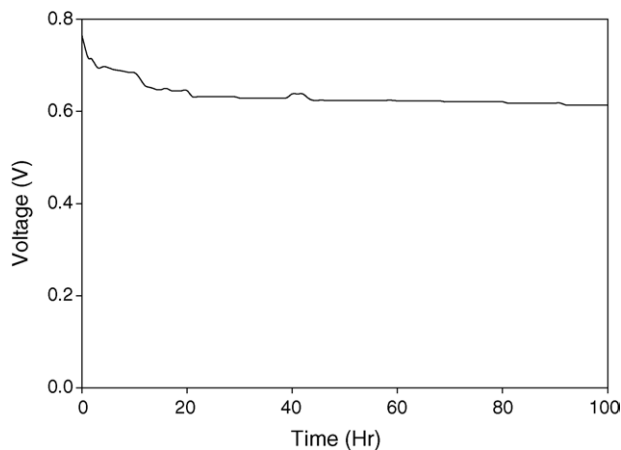


Fig. 3. Long-term test for open circuit voltage (OCV) of the single stack DMFC at 70 °C and 200 kPa.

single stack DMFC for 100 h is shown in Fig. 3. The anode fuel (i.e. liquid methanol) was recycled during this time. The amount of methanol consumed, was compared with the amount of methanol that would have been required if the experiment was conducted with equal flow rate and without recycling. The recycling efficiency was calculated and 97% efficiency was achieved.

The resulting output voltage was compared with the theoretical output, i.e. 1.21 V and the difference can be explained by methanol crossover and contact resistance. Methanol crossover occurs when liquid methanol passes through the MEA and reaches the cathode. The methanol crossover can be reduced by optimizing the concentration of the anode fuel and the thickness of the MEA. The contact resistance of SS 316 increases when an oxide layer is formed on the end plates. The effect can be controlled by optimizing the compaction pressure of the stack.

3.2. Polarization studies

A Polarization study is an electrochemical study to calibrate the performance of the fuel cell, which shows voltage drop as a function of current density. The experiment was carried out at a temperature 70 °C and a pressure 200 kPa. Fig. 4 shows the nature of the voltage and power variation as a function of current drawn from the cell. The stack voltage showed a decrease in nature when the external load current was applied. The test was carried out till the voltage dropped to 0.2 V, which was assumed to be the minimum useful output. A current density of 254 mA cm⁻² was achieved. The power output of the single stack DMFC showed an increase with respect to current density until it reached 210 mA cm⁻² and beyond that the power of fuel stack started decreasing. The meeting point of the voltage and power curve gave the operating point, which is the optimized value of voltage for the single stack of DMFC. The polarization curve indicates about the polarization losses as a function of current density and the

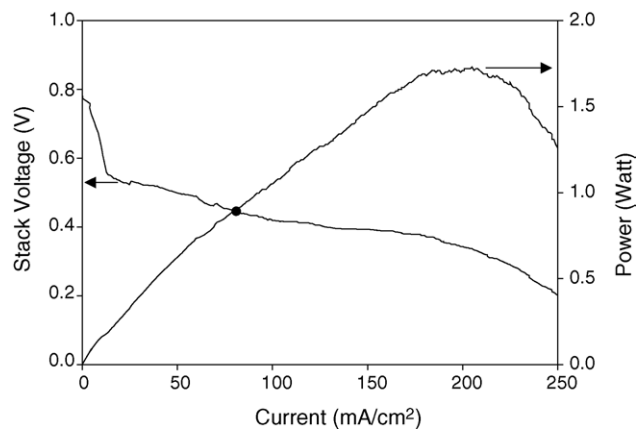


Fig. 4. Performance of single stack DMFC at 70 °C and 200 kPa.

polarization losses can be classified into three types: (1) activation polarization; (2) ohmic polarization; (3) concentration polarization [3]. Activation polarization is the overpotential required to overcome the activation energy of the electrochemical reaction at the catalytic interface. It dominates at lower current density. Activation polarization depends upon the activity of the catalyst used at a particular temperature and can be optimized by adjusting the composition of electrode material. The losses in the linear region refer to ohmic polarization. They include: (1) ionic resistance of the electrolyte; (2) the electrical resistance by end plates; (3) contact resistance between the parts of cell where electrons flow. Ohmic losses can be minimized by improving the electrolyte as a function of temperature and water content. At higher current densities, the losses are dominated by concentration polarization. This can be minimized by proper water and thermal management.

Lower output can be explained by the CO₂ byproduct which forms on the anode and blocks the flow field path thereby reducing the active area. The performance depends on how efficiently the flow field design can drain out the CO₂ from the active area. Heat removal efficiency of flow field has great impact on the performance of the fuel cell. The output of DMFC stack depends largely on the permeability and flow field design of the bipolar plate. The lower the permeability, the better the performance [5,6]. After the long run, the MEA was characterized by the EDS analysis. There were no Fe ions found in the MEA. The analysis showed that there was no dissolution of SS 316 during the experiment.

3.3. Effect of temperature

The polarization of the DMFC was studied at three different temperatures, i.e. 30 °C, 50 °C and 70 °C to monitor the effect of temperature. The pressure was kept constant at 200 kPa. It was found that the polarization curves shifted upward with respect to temperature as indicated in Fig. 5. The performance was better at 70 °C and lower output was received at 30 °C. This can be due to the kinetics of electrochemical reaction involved in the fuel cell, which is temper-

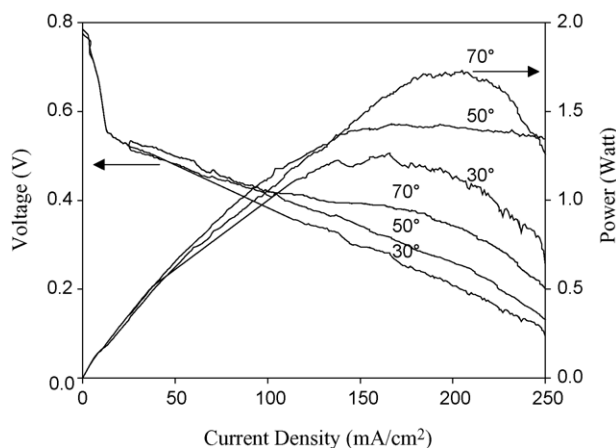


Fig. 5. Variation of voltage and power as a function of temperatures at 200 kPa.

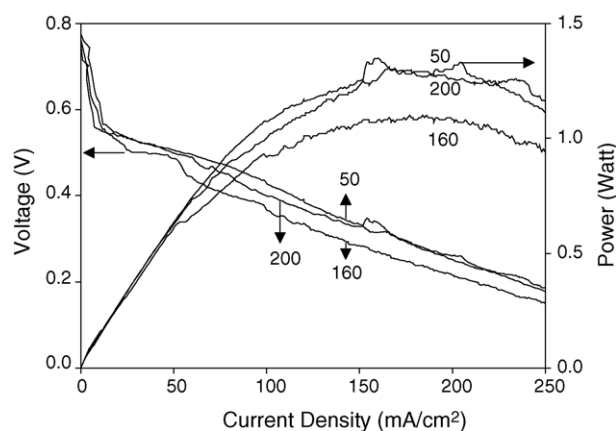


Fig. 6. Variation of voltage and power as a function of pressure at 70 °C.

ature dependent. At higher temperatures, ohmic polarization losses were reduced.

3.4. Effect of pressure

The effect of pressure was studied by conducting the experiments at 70 °C and at different pressures at the cathode. Fig. 6 show the polarization curves at different pressures, i.e. 50 kPa, 160 kPa, 200 kPa. The pressure of the cathode gas was changed and the performance was monitored. The test was done by keeping anode pressure constant at atmospheric pressure. At 50 kPa, there was better performance than at 160 kPa. However, it was found that after 160 kPa, increase of pressure shifted the polarization curve upward. A better performance was achieved at 200 kPa as compared with 160 kPa. It was because reactant partial pressure, gas solubility and mass transfer rate an increased at higher pressure [6].

4. Conclusions

A single stack DMFC was developed with SS 316 end plates. Experiments were carried out with a modified serpentine design on SS 316 end plates. A long-term test was carried out at 70 °C and 200 kPa on the single stack DMFC and a consistent open circuit voltage of 0.614 V was achieved. A Polarization test was done at 70 °C and 200 kPa and a current density of 254 mA cm⁻² was achieved. Experiments were conducted at different temperatures and pressures to optimize the operational parameters. The results indicate that the best performance of the single stack DMFC developed at our laboratory was achieved at 70 °C and 50 kPa. The present investigations suggest that SS 316 can be an alternative bipolar plate material for DMFC. It satisfies DOE's target regarding the cost, weight, volume, properties of bipolar plate material and will reduce the machining cost, as well as the volume of fuel cell stack.

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References

- [1] A.K. Shukla, C.L. Jackson, K. Scott, R.K. Raman, *Electrochim. Acta* 47 (2002) 3401–3407.
- [2] M. Baldauf, W. Preidel, *J. Power Sources* 84 (1999) 161–166.
- [3] *Fuel Cell Handbook*, sixth ed., EG&G Technical Services Inc., US Department of Energy, Office of Fossil Energy, National Energy Technology Laboratory, West Virginia, 2002, pp. 3.16–3.17.
- [4] *Automotive Propulsion Materials*, US Department of Energy, Energy efficiency and renewable energy, Office of Transportation Technologies, FY 2001 progress report (http://www.ornl.gov/sci/apm/documents/2001_pr_propulsion_materials.pdf May 11, 2005).
- [5] A. Kumar, R.G. Reddy, *J. Power Sources* 114 (2003) 54–62.
- [6] A. Kumar, R.G. Reddy, *J. New Mater. Electrochem. Syst.* 6 (2003) 231–236.
- [7] A. Kumar, R.G. Reddy, in: D. Chandra, R.G. Baustia (Eds.), *Fundamental of Advanced Materials for Energy Conversion*, TMS, Warrendale, 2002, pp. 41–53.
- [8] A.S. Woodman, K.D. Jayne, E.B. Anderson, M.C. Kimble, Development of corrosion-resistant coatings for fuel cell bipolar plates, in: *AESF SUR/FIN '99 Proceeding*, vol. 6, American Electroplater and Surface Finishers Society, 1999, pp. 21–24.
- [9] J. Wind, R. Spah, W. Kaiser, G. Bohm, *J. Power Sources* 105 (2002) 256–260.
- [10] H. Yang, T. S. Zhao, *Electrochim. Acta*, in press.
- [11] A.S. Arico, P. Creti, V. Baglio, E. Modica, V. Antonucci, *J. Power Sources* 91 (2000) 202–209.