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Journal of Power Sources 114 (2003) 195-202

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# Improved membrane and electrode assemblies for proton exchange membrane fuel cells

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Received 19 October 2002; accepted 25 October 2002

#### Abstract

Three electrodes—E1 (0.18 mg Pt cm<sup>-2</sup>), E2 (0.38 mg Pt cm<sup>-2</sup>), E3 (0.4 mg Pt cm<sup>-2</sup> without a gas-diffusion layer)—are fabricated and compared with a commercial product (E-TEK). The performance of the electrodes increases with increase in Pt loading in the catalyst layer. The performance of the E2 electrode is superior to that of E1, E-TEK or E3. Elimination of the diffusion layer between the carbon-cloth substrate and the catalyst layer affects the performance of electrode E3 in particular. The power density shows a similar pattern to current density. The difference in performance between E2 and E-TEK electrodes may be due to the difference in the method of fabrication. Increase in exchange current density results in an increase in efficiency. The curves for E1, E2 and E-TEK electrodes appear to stabilize at constant efficiency, which indicates maximum efficiency at a lower exchange current density, compared with the E2 electrode, which does not approach a steady efficiency even at an exchange current density of 1 mA cm<sup>-2</sup>. This means that the E2 electrode has greater efficiency than E1, E3 or E-TEK electrodes. Voltage and irreversibility curves for the four electrodes meet at different voltage operational limits; namely, 0.48, 0.55, 0.46 and 0.42 V at 1.2, 0.85, 0.4 and 0.3 mA cm<sup>-2</sup>, for E-TEK, E2, E1 and E3 electrodes, respectively. Hence, while these electrodes can be operated conveniently, the E2 electrode with a 0.38 mg Pt cm<sup>-2</sup> loading can be operated at optimum conditions of 0.55 V and 0.85 mA cm<sup>-2</sup>.

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Keywords: Fuel cell; Membrane and electrode assembly; Hydrogen; Irreversibility; Optimization; Platinum catalyst

## 1. Introduction

The proton exchange membrane (PEM) fuel cell is one of the presently known five types of fuel cell that convert chemical energy of fuels (e.g. H<sub>2</sub> and O<sub>2</sub>) directly into electricity, heat and water. The PEM fuel cell is commonly claimed to be the most promising technology due to its portable power and residential applications. The membrane and electrode assembly (MEA) is often stated to be the heart of the PEM fuel cell. The MEA consists of a sheet of proton-conducting polymer electrolyte membrane with two electrodes—the negative (anode) and positive (cathode)—bonded to the opposite sides of the sheet. The arrangement is then compressed on both sides by grooved bipolar plates, or grooved end-plates in the case of a single cell, to transport

the  $H_2$  and  $O_2$ /air, respectively, to the electrodes. In this study, the electrodes are fabricated with 10 wt.% platinum, which is supported on carbon black, as the electrocatalyst 60 wt.% polytetrafluoroethylene (PTFE) as a binder and wet-proofing agent, and Nafion® solution as a proton conductor between the catalyst layer and the Nafion® (117 mm) membrane.

To maximize the output of the fuel cell, it is important that every catalyst particle has an electronically conductive path to the flow field, an ionically conductive path to the electrolyte membrane, and a path for gas access. Unfortunately, the type of porous structure required to achieve such output is not consistent with the gas-barrier properties needed for the bipolar plates and end-plates of the fuel cell. To combat such inconsistency, a porous and conductive gas-diffusion layer is required to ensure good electrical contact and good gas access to all parts of the electrocatalytic layer. The gas-diffusion layer should also be mechanically strong and

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resistant to acid media and humidity. Moreover, it functions as a physical barrier and hence prevents the loss of the catalytic layer from the MEA. The conversion efficiency of the MEA depends on many factors that include type and thickness of the gas-diffusion material, the Nafion<sup>®</sup> to carbon ratio in the catalysts, and the method used for MEA preparation. The failure modes of PEM fuel cells are likely to be low-level contamination, corrosion of the plates leading to increased contact resistance, thermal or hydration cycling leading to mechanical stress, catalyst particle ripening, and swelling of polymer materials in the active catalyst layer. Other problems include waterremoval characteristics, compaction of the gas-diffusion layer due to mechanical stresses, and surface chemistry changes in the gas-diffusion layer. Thus, irreversibility analysis is a useful tool for product development of both MEAs and stacks as a whole. In this study, 10 wt.% platinum-supported carbon and a 60 wt.% PTFE mixture were cast on carbon cloth and coupled into two types of MEA—one with a gas-diffusion layer, the other without a gas-diffusion layer—to evaluate the irreversibility of the transformation from chemical energy to electrical energy, heat and water.

# 2. Theory

In the operation of the PEM fuel cell, hydrogen fuel is oxidized at the anode, and oxygen is reduced at the cathode to produce electricity, heat and water, according to the balance of inputs and outputs shown in Fig. 1. The electrochemical combination of these fuels and their overall reaction can be represented, respectively, as:

$$H_2|H^+, H_2O|O_2$$
 and  $2H_2 + O_2 \rightarrow 2H_2O$  (1)

where the vertical lines represent the phase change from hydrogen gas to dissolved ions in liquid water and then to oxygen gas on the reverse side. The oxygen electrode has a higher potential than the hydrogen electrode, hence oxygen is reduced to water and hydrogen is oxidized to H<sup>+</sup> when the electrodes are connected externally. Oxygen reduction has been the focus of extensive research due to its technological importance in fuel cells and metal–air batteries. Larminie

and Dicks [1] have emphasized that in low- and mediumtemperature fuel cells activation overvoltage is the most important irreversibility and cause of voltage drop, which occurs at the oxygen-reducing cathode. In fact, the reaction at the oxygen electrode undergoes an equilibrium stage where there is a continual backward and forward flow of electrons from and to the electrolyte, as expressed by:

$$O_2 + 4e^- + 4H^+ = 2H_2O$$
 (2)

Thus, for a PEM fuel cell that has no losses except for the irreversibility on the oxygen electrode, the voltage may be given by Eq. (3):

$$V = E - A \ln \left(\frac{i}{i_0}\right) \tag{3}$$

where  $A = RT/2\alpha F$ ,  $\alpha$  is the charge-transfer coefficient, which is proportional to the electrical energy applied in changing the rate of an electrochemical reaction, F the Faraday constant, R the gas constant, T the absolute temperature, i the current density, and  $i_0$  the exchange current density at which the irreversibility (right-hand term in Eq. (3)) begins to move from zero. Eq. (3) can be rearranged into:

$$V = E - A \ln i - Irr_0 \tag{4}$$

where  $Irr_0 = A \ln i_0$  and is termed as the 'exchange irreversibility', and E the reversible open-circuit voltage. It is obvious from Eqs. (3) and (4) that  $i_0$ , and eventually  $Irr_0$  is crucial in controlling the performance of a fuel-cell electrode. The smaller the  $i_0$ , the greater is the irreversibility and, thus, the greater is the voltage drop of the cell. Hence, if the exchange current density,  $i_0$ , is sufficiently high, the surface area of the electrode is more active and results in current flow in one particular direction of Eq. (2).

# 3. Experimental

The experimental work involved electrode preparation, treatment of the Nafion membrane, membrane and electrode assembly, single-cell assembly, and operation of single-PEM fuel cell [2]. The experimental set-up, MEA and mode of operation are shown schematically in Fig. 2.

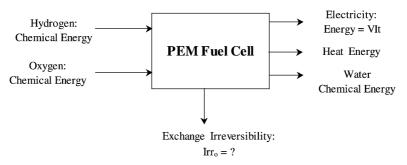


Fig. 1. Schematic of balance of fuel cell inputs and outputs.

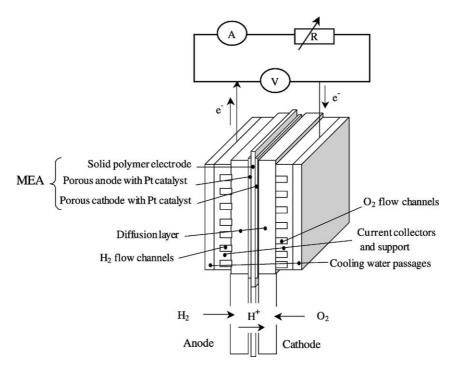


Fig. 2. Schematic of MEA and single-cell testing apparatus.

## 3.1. Electrode preparation

Carbon cloth 'A' (E-TEK Inc.) with a thickness of 350 µm was used as a substrate for casting of a platinum-supported carbon, 10 wt.% Pt/C (Fluka, Chemie AG), activated carbon black (Ajax Chemicals, Sydney), and Teflon (PTFE) 60 wt.% (Aldrich) mixture. Three different structures of electrode were prepared with 70 wt.% Pt/C and 30 wt.% Teflon [3]. One structure was with a diffusion layer 0.18 mg Pt cm<sup>-2</sup> loading, designated E1; the second was with a diffusion layer and 0.38 mg Pt cm<sup>-2</sup> loading, designated E2; while the third was without a diffusion layer but with 0.4 mg Pt cm<sup>-2</sup> loading.

Three layers are known to make up a typical electrode [4], namely, the backing layer (carbon cloth and Teflon), the diffusion layer (carbon black and Teflon), and the catalyst layer (platinum, carbon and Teflon). The following simple process was used to fabricate electrodes with different low loadings of platinum. The backing layer was prepared by applying Teflon emulsion on to the carbon cloth. This was achieved by first making the carbon-cloth hydrophobic by impregnation with 15 wt.% Teflon using 60 wt.% Teflon solution for 3–4 min. The cloth was dried in an oven at 80 °C to remove solvent water. The backing layer and diffusion layer were in contact and had a total thickness of 350 µm, which was measured after impregnating the carbon cloth with 30 wt.% Teflon solution. The diffusion layer was prepared from carbon black with 30 wt.% Teflon using 60 wt.% PTFE, which was mixed with water and alcohol as solvent, using magnetic stirrer, for a few hours. The prepared ink was applied on the backing layer by means of a spraying method. It was then dried at 80 °C for 1 h to

remove the residual water and alcohol. The diffusion layer with a loading of 0.4 mg Pt cm<sup>-2</sup> was in contact with the catalyst layer. This loading corresponded to a thickness of 40 μm. The catalyst was prepared from platinum-supported carbon black 10% Pt/C and 30 wt.% Teflon. First, a homogenous suspension was made by stirring platinum supported by the carbon black and Teflon with isopropyl alcohol as solvent. The mixing took a few hours until a homogenous solution was obtained. The ink obtained was applied to the diffusion layer (PTFE/C) with a spraying method and slurry casting, and then dried at 80 °C for 1 h to remove the solvents, water and alcohol. Finally, the electrode was baked at 200 °C for 20 min in an oven, to remove the residual surfactant in the catalyst layer, followed by sintering at 280 °C for 20 min. The thickness of the catalyst layer is dependent on the loading of platinum-supported carbon. The catalyst layer can be described as a Pt/C/ionomer composite, where each of the three components is distributed uniformly within the volume of the layer. The E1, E2 and E3 electrodes were all dried at 80 °C to remove water and isopropyl alcohol. This was followed by thermal treatment at 200 °C to remove the dispersion agent contained in PTFE, and later sintered at 280 °C for 20 min. This thickness of this layer typically varies between 10 and 40 µm.

## 3.2. Nafion membrane treatment

Nafion<sup>®</sup> (117 mm) membrane sheet of  $25 \text{ cm}^2$  was first cleaned with distilled water in a water bath (Memmer, Germany) at 80–90 °C, followed by heating in 5% H<sub>2</sub>O<sub>2</sub> (J.T. Baker Inc., USA) for 1 h at 70–80 °C to remove organic impurities. It was then heated in 0.5 M H<sub>2</sub>SO<sub>4</sub> (J.T. Baker

Inc., USA) for 1 h at 70–80 °C. The  $H_2SO_4$  was removed by repeated washing in boiling water [5]. The membrane was stored in the dark overnight in distilled water before assembly with the electrodes.

## 3.3. Membrane and electrode assembly (MEA)

The final preparation of electrodes with platinum loadings of 0.18, 0.38 or 0.4 mg Pt cm $^{-2}$  and impregnated with a 5% Nafion $^{\circledR}$  solution was achieved by means of a brushing technique [6,7]. The electrodes were air dried at 80 °C and then weighed. The electrodes and the membrane were coupled into a single-cell MEA by a hot pressing procedure [6] using a press (Scientific Press-Motorized Hydraulic 20 tonnes) at a temperature of 120 °C and a pressure of 1000 psi for 90 s. The MEA was then cooled at room temperature for 30 min.

# 3.4. Single-cell PEM fuel-cell assembly

The major components of the single-cell assembly are two graphite plates, the MEA, gaskets containing the gas inlet and outlet, and ribbed or grooved channels for distribution of the reactant gases behind the porous gas-diffusion electrodes. The MEA was positioned between the two graphite plates with the Teflon coated gasket placed between the membrane and each of the graphite plates to prevent gas leakage, and to avoid excessive compression of the electrodes. Copper plates were used as current-collectors and were positioned behind the graphite plates. Two, thick, stainless-steel end-plates with bolts were used to compress the overall assembly, with Teflon sheets for electric insulation placed between the single cell and the end-plates.

## 3.5. Operation of single cell in fuel-cell test apparatus

During operation of PEM fuel cells, the following processes take place within the electrode (Fig. 2): (i) the reactant gases diffuse through the porous backing layer; (ii) at the gaslelectrolyte interface, the gases dissolved and then diffuse to the electrolytelelectrode interface; (iii) electrocatalytic reaction inside the catalyst layer precedes the gas adsorption at the electrode surface; (iv) ionic transport occurs in the electrolyte, but electronic transport takes place in the electrode.

The single cell was installed in a fuel-cell testing apparatus (Fig. 2) equipped with gas sources, temperature control, and gas flow-rate control using rotameters (Cole Palmer Instrument Company), back-pressure regulators for hydrogen and oxygen (Gas-Arc Multi-stage), and a load box resistance type KF19 (Lionmount and Co. Ltd.). Oxygen, hydrogen and compressed air supplied by Malaysian Oxygen Berhad (MOX) were used in the fuel-cell operations. Hydrogen was passed through a humidifier to wet the gas, and fed into the anode at a flow rate of 140 ml min<sup>-1</sup> and 1 atm. Oxygen (some times air) entered the fuel cell through the cathode at a flow rate of 380 ml min<sup>-1</sup> and 2 atm. The electrons generated from the anode were connected to a digital multimeter (1906 Competing Multimeter), with an external variable resistance, as shown in Fig. 2 to measure the current and voltage produced by the cell.

#### 4. Results and discussion

Activation losses are often encountered in operating fuel cells. These losses are caused by the slow rate of reactions on the surface of the electrodes, while a portion of the voltage generated is lost in driving the chemical reaction that results in the electrons migrating to or from the electrode, which, in turn, results in irreversibility. The entire magnitude of irreversibility in running the fuel cell contains one major component. This is referred to as the exchange irreversibility (Irr<sub>0</sub>) and is derived from its constituent, the exchange current density,  $i_0$  (Eq. (4)). These parameters are examined in this study.

## 4.1. Single-cell PEM fuel-cell performance

The characteristics of the fabricated electrodes and the commercial electrode are listed in Table 1 in terms of Pt/C loading, reversible open-circuit potential, power, and resistance. The open-circuit potential for electrode E2 decreases progressively with time and reaches a steady-state value within 2.5 h with hydrogen as fuel and oxygen or air as the reactant. The single-cell operating with pure oxygen gave a better performance at a potential 0.912 V than that using air at potential of 0.854 V (Fig. 3).

Potential versus current density curves for different electrode structures with varying Pt loading are shown in Fig. 4.

Table 1 Characteristics of fabricated and commercial electrodes

Electrode	Catalyst loading (mg Pt cm <sup>-2</sup> )	Open-circuit voltage (V)	Power (mW cm <sup>-2</sup> )	Resistance $(\Omega \text{ cm}^{-2})$
E1	0.18	0.937	4.44	0.05
E2	0.38	1.077	21.57	0.01
E-TEK (commercial)	0.4	0.989	20.32	0.012
E3 (without diffusion layer)	0.4	0.934	2.94	0.064

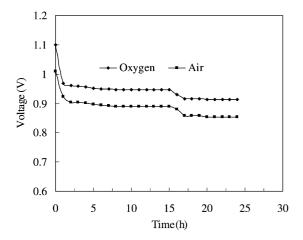


Fig. 3. Performance kinetics of single-cell operating with  $O_2$  or air using  $H_2$  as fuel with 0.38 mg Pt cm<sup>-2</sup> loading (electrode E2).

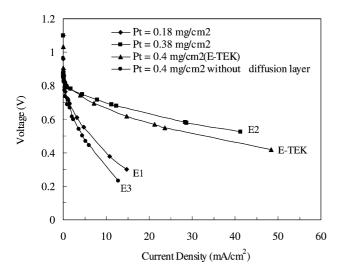


Fig. 4. Cell potential as function of current density for a single cell  $(25 \, \mathrm{cm}^2)$  for electrodes with varying Pt loadings and operating at  $25 \, ^{\circ}\mathrm{C}$  with a  $\mathrm{H_2:O_2}$  ratio of 1:2 using Nafion  $^{\circledR}$  117 solid membrane electrolyte.

The performance of the electrodes increases with increase in Pt loading in the catalyst layer when made with the same fabrication process and run at the same operating conditions. The E-TEK electrode has a much better performance than the E3 electrode, which has a similar 0.4 mg Pt cm<sup>-2</sup> loading but does not have a gas-diffusion layer. The E2 electrode with 0.38 mg Pt cm<sup>-2</sup> delivers a superior performance compared with the others (E1, E-TEK and E3). It is apparent that elimination of the diffusion layer between the carbon-cloth substrate and the catalyst layer influences the performance of electrode E3 compared with the other electrodes. The current density of E3 falls about four times faster than that of the E-TEK electrode with the same platinum loading. This is perhaps due to the inaccessibility of the electrons to diffuse freely in the E3 electrode that has no diffusion layer. The slope of the linear section of the current density plot, which is the ohmic resistance, R, increases from  $0.012 \,\Omega \,\mathrm{cm}^{-2}$  for the E-TEK electrode to  $0.064~\Omega~cm^{-2}$  for the E3 electrode. This means that in an electrode without a gas-diffusion layer, the current losses are about four times greater than in E-TEK and E2. Conversely, increase in the platinum loading in electrodes with diffusion layers increases the performance as demonstrated by the better performance of the E2 electrode with 0.38 mg Pt cm<sup>-2</sup> compared with that of the E1 electrode with 0.18 mg Pt cm<sup>-2</sup>. The linear slope of E2  $(0.1 \,\Omega \,\mathrm{cm}^{-2})$  is five times less than that of E1  $(0.05 \,\Omega\,\mathrm{cm}^{-2})$ . Wilson et al. [8] also concluded that diffusion of the microporous carbon cloth and the catalyst layer, as in the case of the E2 electrode, reduces the amount of water droplets formed at the cathode and, thereby, improves electrode performance. The location of the electrocatalyst particles in the carbon-cloth pores due to the hot pressing of E3 electrode is such that there is a low density of electrocatalyst sites near to the membrane electrolyte. On the other hand, the microporous carbon diffusion layers in both El and E2 electrodes are used to support the density of the

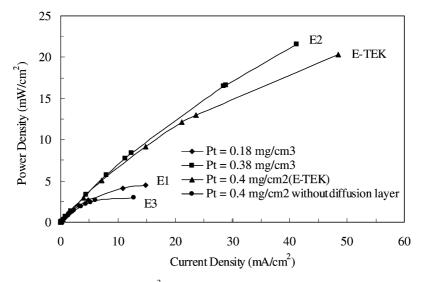


Fig. 5. Power density vs. current density for a single cell  $(25 \text{ cm}^2)$  for electrodes with varying Pt loadings and operating at  $25 \,^{\circ}\text{C}$  with a  $\text{H}_2:\text{O}_2$  ratio of 1:2 using Nafion<sup>®</sup> 117 solid membrane electrolyte.

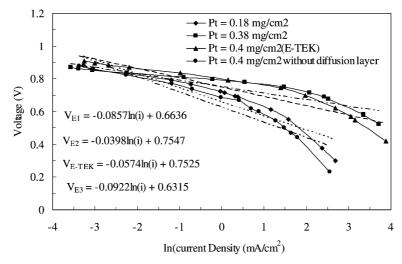


Fig. 6. Voltage vs. ln i in Eq. (4) (broken lines are linear correlation lines) for electrodes E1, E2, E3 and E-TEK.

electrocatalyst. Perhaps, therefore, the density of the eletrocatalyst sites are higher in E1 and E2 than in E3 which results in an improved electrocatalyst–membrane electrolyte contact. Such contact may lead to the improved performance. A similar observation has been made by Passalacqua et al. [9].

The power density of the electrodes as a function of current density is shown in Fig. 5. The power density increases with Pt loading. The power density increases from 4.44 mW cm<sup>-2</sup> for the E1 electrode to 21.57 mW cm<sup>-2</sup> for the E2 electrode. The power density increases from

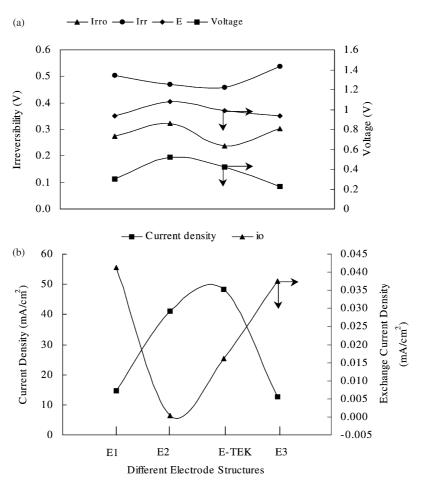


Fig. 7. Effect of electrode structure on: (a) irreversibility and cell voltage; (b) current density and exchange current density.

2.94 mW cm<sup>-2</sup> for the E3 electrode to 21.57 mW cm<sup>-2</sup> for the E2 electrode with a diffusion layer. Fig. 5 also shows that the E2 electrode gives a better performance than the E-TEK electrode with a slightly greater Pt loading. The power density for the E2 electrode is 21.57 mW cm<sup>-2</sup>, while that for the electrode is 20.57 mW cm<sup>-2</sup>. The difference in performance between the E2 and E-TEK electrodes may be due to the difference in the methods used for their fabrication, which may result in different particle distributions of the electrocatalyst.

## 4.2. Irreversibility in activation losses

The voltage obtained from the single cell is plotted against ln i according to Eq. (4) in Fig. 6 for the four electrodes. The parameters obtained from the lines of best fit have been substituted into Eq. (4) and the total irreversibility (Irr) and exchange irreversibility (Irr<sub>0</sub>) due to activation losses have been determined. The effect of the different electrode structures on irreversibility and voltage is demonstrated in Fig. 7(a), and the effect of different electrode structures on current density (i) and exchange current density  $(i_0)$  are given in Fig. 7(b). It can be seen that the values of exchange irreversibility, Irr<sub>0</sub>, and exchange current density,  $i_0$  are much smaller than the respective values of irreversibility, Irr and i. This agrees with the relationship between Eqs. (3) and (4). These results also agree with the literature [1] in terms of the relationship between i and  $i_0$ , i.e. that the former must be greater than the latter for Tafel equation to be true, and that the irreversibility (overvoltage) is:

$$Irr = A \ln \left( \frac{i}{i_0} \right) \tag{5}$$

The data in Fig. 7(a) also confirmed the basic truth that the voltage obtained is much smaller than the open-circuit

potential, and that the electrode structures (i.e. E1, E2, E-TEK and E3) exert a marked effect on the achievable performance and the irreversibility. This implies that, to expect a good output from a PEM fuel cell, the method of electrode fabrication is very important. Thus, it can be concluded that the electrode fabricated with a loading of 0.38 mg Pt cm<sup>-2</sup>, viz. E2, gives the best performance and almost the least irreversibility. On the other hand, since the exchange current density is the parameter responsible for the control of the electrode performance, the results in Fig. 7(b) clearly demonstrate that the electrode with structure E2 (0.38 mg Pt cm<sup>-2</sup>) has the least exchange current density and also shows an appreciative current density. Nevertheless, its current density is slightly less than that of the E-TEK electrode, and therefore some improvements are still required in its fabrication processes.

## 4.3. Electrode efficiency and operational limits

The stoichiometric ratio of  $H_2$ : $O_2$  in PEM fuel-cell operation is 2:1, see Eq. (1). This means that exactly 2 mol of hydrogen are required by 1 mol of oxygen to produce 2 mol of water. In practice, however, the hydrogen passed into the fuel cell is not completely used, some always has to pass through. Therefore, single-cell efficiency is given by Larminie and Dicks [1] as:

efficiency (%) = 
$$\mu_{\rm f} \frac{V}{1.48} \times 100$$
 (6)

where

$$\mu_{\rm f} = \frac{{\rm mass~of~fuel~reacted~in~cell}}{{\rm mass~of~fuel~input~to~cell}}$$

The relationship between the single-cell efficiency and the exchange current density is presented in Fig. 8(a). The data show that an increase in exchange current density

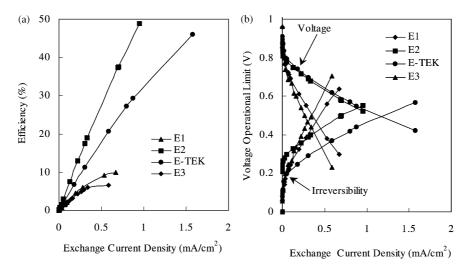


Fig. 8. Effect of exchange current density on: (a) cell efficiency; (b) voltage and electrode irreversibility. Data used to determine voltage operational limit for single cell with different electrode structures.

results in an increase in efficiency. The curve for the E2 electrode had the steepest slope followed by those for the E-TEK, E1 and E3 electrodes. The curves for the latter three electrodes appear to stabilize at a constant efficiency, which indicates that their maximum efficiency is at a lower exchange current density compared with the E2 electrode, which does not approach a steady efficiency even at an exchange current density of 1 mA cm<sup>-2</sup>. This means that the E2 electrode has a greater efficiency than the E1, E3 and E-TEK electrodes. The voltage operational limit in terms of voltage and irreversibility is given in Fig. 8(b) as a function of the exchange current density. The voltage and irreversibility curves for the four electrodes meet at different voltage operational limits. For electrodes E-TEK, E2, E1 and E3, the voltage operational limits are 0.48, 0.55, 0.46 and 0.42 at 1.2, 0.85, 0.4 and 0.3 mA cm<sup>-2</sup>, respectively. Hence, while these electrodes can be operated conveniently, the E2 electrode with a 0.38 mg Pt cm<sup>-2</sup> loading possesses the optimum voltage operational limits.

## 5. Conclusions

Three different structures of electrodes have been prepared with 70 wt.% Pt/C and 30 wt.% Teflon. The electrode with a 0.18 mg Pt cm<sup>-2</sup> loading is designated E1 and that with a 0.38 mg Pt cm<sup>-2</sup> loading is designated E2. A third electrode, E3, does not have a diffusion layer, but has a 0.4 mg Pt cm<sup>-2</sup> loading. The fourth electrode, E-TEK, is a commercial product. The performance of the electrodes increases with increase in Pt loading in the catalyst layer and the E2 electrode shows the best performance. It is apparent that the elimination of the diffusion layer between the carbon-cloth substrate and the catalyst layer affects the performance of the E3 electrode. This is perhaps due to the inaccessibility of the electrons to diffuse freely in the electrode. The slope of the linear section of the current density plot, which represents the ohmic resistance, increases from  $0.012\,\Omega\,\text{cm}^{-2}$  for the E-TEK electrode to  $0.064~\Omega~\text{cm}^{-2}$  for the E3 electrode. Thus, in the absence of a gas-diffusion layer, the current losses are about four times greater. The linear slope of E2 (0.1  $\Omega$  cm<sup>-2</sup>) was five times less than that of E1 (0.05  $\Omega$  cm<sup>-2</sup>). It is also observed that the power density increased from 4.44 mW cm<sup>-2</sup> for the E1 electrode to 21.57 mW cm<sup>-2</sup> for the E2 electrode. The power density increases from 2.94 mW cm<sup>-2</sup> for the E3 electrode to 21.57 mW cm<sup>-2</sup> of the E2 electrode with the diffusion layer. By contrast, the power density of the E-TEK electrode is 20.57 mW cm<sup>-2</sup>. This difference in performance between the E2 and the E-TEK electrodes may be due to the difference in the method of fabrication of the electrodes. An increase in exchange current density results in an increase in efficiency. The curve for the E2 electrode has the steepest slope, followed by E-TEK, E1 and E3 electrodes. The curves for the latter three electrodes tend to stabilise at constant efficiency, which suggests that their maximum efficiency is at a lower exchange current density than that of the E2 electrode, which does not approach a steady efficiency even at an exchange current density of  $1 \text{ mA cm}^{-2}$ . This means that the E2 electrode has a greater efficiency than the E1, E3 and E-TEK electrodes. The voltage and irreversibility curves for the four electrodes meet at different voltage operational limits. For the E-TEK, E2, E1 and E3 electrodes, the voltage operational limits are 0.48, 0.55, 0.46 and 0.42 V at 1.2, 0.85, 0.4 and 0.3 mA cm<sup>-2</sup>, respectively. This means that the E2 electrode possesses voltage operational limits for a single cell.

## References

- [1] J. Larminie, A. Dicks, Fuel Cell Systems Explained, Wiley, Chichester, 2001, p. 17.
- [2] C. Rachid, Fabrication of Low Platinum Loading Electrode for Proton Exchange Membrane Fuel Cells System (PEMFCs), Masters Thesis, Universiti Kebangaan Malaysia, Malaysia, 2001, p. 43.
- [3] S. Mukerjee, S. Srinivasan, A.J. Appleby, Electrochim. Acta 38 (1993) 1661.
- [4] K.G. Sasi, M. Raja, S. Parthasarathy, Electrochim. Acta 40 (1994) 285.
- [5] A.T. Zawodzinski, C. Derouin Jr., S. Radzinski, R.J. Sherman, W.T. Smith, T.E. Springer, S. Gottesfed, J. Electrochem. Soc. 140 (1993) 1041.
- [6] E.A. Ticianelli, C.R. Derouin, A. Redondo, S. Srinivasan, J. Electrochem. Soc. 135 (1988) 2209.
- [7] N.B. Felix, S. Srinivasan, J. Electrochem. Soc. 144 (1997) 2767.
- [8] M.S. Wilson, J.A. Valerio, S. Gottesfeld, Electrochim. Acta 40 (1995) 355
- [9] E. Passalacqua, F. Lufrano, G. Squadrito, A. Patti, L. Giorgi, Electrochim. Acta 43 (1998) 3665.