

# Power loss and its effect on fuel cell performance

M. Shen\*, K. Scott

*School of Chemical Engineering & Advanced Materials, University of Newcastle upon Tyne, Newcastle upon Tyne NE1 7RU, UK*

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## Abstract

Fuel cell performance and the power are influenced by factors referred to as “power loss”. In fuel cells, there are two kinds of power losses: one is dominated by the electric resistance between the electrodes, which is called as leak resistance, and the other is dominated by mass diffusion between the anode and cathode, i.e. “crossover”. In this work, we analyse the two kinds of power losses and discuss how they influence fuel cell performance. The power loss of a fuel cell caused by crossover is described by a new parameter  $P_{\text{leak}}$ . The practical performance curve of direct methanol fuel cells using different types of membrane materials are modelled by a mathematical equation describing the power loss and crossover effect. This equation is used to estimate the methanol crossover flux.

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**Keywords:** Fuel Cell; Power loss; Crossover; Leak resistance; Membrane; DMFC

## 1. Introduction

Fuel cells are promising candidates for portable power and transportation applications, due to their higher energy efficiency, energy density and very low emissions. Polymer electrolyte fuel cells (PEFC) systems are of growing interest because of their low operating temperature and high export power density. The  $\text{H}_2/\text{O}_2$  PEFC has experienced considerable progress during recent years [1]. Compared to the  $\text{H}_2/\text{O}_2$  PEFC, another kind of PEFC, the direct methanol fuel cell (DMFC) has advantages of easier fuel delivery and storage, no humidification requirement and simpler design [2].

In fuel cells, power loss is a very important factor influencing performance. There are two kinds of power loss: one is dominated by the electric resistance between the electrodes, which is called as leak resistance, and the other is dominated by mass diffusion between the anode and cathode, i.e. crossover. Methanol crossover is considered to be one of the biggest problems for the direct methanol fuel cells [3,4]. Various methods have been developed to mea-

sure the crossover rate in fuel cell operation. Tricoli et al. [5] have investigated the methanol permeability in two partially fluorinated ionomeric commercial membranes (manufactured by Pall) which showed lower methanol permeation than Nafion® membrane, making them potential membranes for the DMFC.

Ren et al. [6,7] measured the flux across Nafion® membranes during DMFC operating. The methanol crossover rate was studied with various concentrations of methanol solution. The diffusion coefficient and methanol concentration in the membrane have been determined from the measured transient limiting current density, following a potential step.

Okada et al. [8] have studied the ion and water transport characteristics of Nafion® membrane and found that the mobility of the ions, the interaction of the ions with water and microscopic membrane channel structures are important factors. They also investigated the electrostatic effect between the ion and water dipole and the size effect of the cation, which influences the water transfer coefficient.

Recently, new techniques have been developed [9–11] to determine the methanol permeation in DMFC operation. The results [10] show clearly that the methanol permeation rate decreases as operating current increases. Methanol diffusion

\* Corresponding author.

E-mail address: [mu-zhong.shen@newcastle.ac.uk](mailto:mu-zhong.shen@newcastle.ac.uk) (M. Shen).

coefficients in the membrane have also been estimated from open circuit potential measurements in direct methanol fuel cells [12].

Quantitative models can be valuable in the interpretation of experimental observation and in the development and optimization of the system. Cruickshank and Scott [13] identified and analysed the effect of methanol crossover on DMFC performance. They developed a simple model for methanol transport through the membranes and its effects on cathodes overpotential. Gurau and Smotkin [14] also attempted to relate methanol crossover in DMFC to power and energy densities.

Other mathematical models equations have been used to describe DMFC fuel cells [15] and also other kind of fuel cells [16]. The fundamental principle of the above is based on the calculation of the potential of fuel cell with the equation:

$$V_{\text{cell}} = E_{\text{cell}} - \eta_{\text{an}} - \eta_{\text{cat}} - \eta_{\text{ohmic}} - \eta_{\text{xover}} \quad (1)$$

where  $E_{\text{cell}}$  is equilibrium potential of fuel cell,  $\eta_{\text{an}}$  and  $\eta_{\text{cat}}$  are the overpotentials at the anode and cathode,  $\eta_{\text{ohmic}}$  is the ohmic overpotential which is calculated for the internal resistance of fuel cell,  $\eta_{\text{xover}}$  is the overpotential caused by methanol crossover.

Present modelling of the DMFC attempts to precisely define all overpotential values. For different kinds of fuel cell and/or different operating condition, the overpotential values are different. A good modelling result will involve many empirical parameters, which may only suit specific operating condition and a specific fuel cell design. To avoid using too many empirical parameters in evaluating fuel cell performance, here we use the regulation [17] of power converted from chemical energy to electrical energy as a fundamental principle to analysis power loss of a fuel cell. This regulation is described as: total power produced is proportional to the square of the potential difference between the equilibrium potential and work potential. With this regulation, we derive a mathematical equation to describe the performance curve of general chemical power sources with three power source parameters:  $E_0$ , equilibrium potential;  $R$ , internal resistance;  $K$ , power conversion coefficient. The values of parameters calculated from this equation agree with performance curves of practical chemical power sources, such as different type of fuel cells and batteries in operating conditions. In this paper, we discuss the fuel cell performance and its mathematical expression when the power losses are dominated by the leak resistance between the electrodes and fuel, e.g. methanol crossover.

## 2. Experimental details

The DMFC, shown schematically in Fig. 1, had a cross-sectional area of  $9 \text{ cm}^2$ . The cell was fitted with one membrane electrode assembly (MEA) sandwiched between two graphite blocks each of which had flow beds, in the form of parallel channels, for methanol or oxygen/air flow. The cell

was held together with two plastic insulation sheets and two stainless steel 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 cell was used in a simple flow rig, which consisted of a Watson Marlow peristaltic pump to supply aqueous methanol solution, from a reservoir, to a water bath, which keeps the solution at a constant temperature. Air was supplied from cylinders, at ambient temperature, and the pressure regulated by pressure regulating valves.

MEAs studied in this work were made in the following manner: the anode consisted of a teflonised (20%) carbon paper (E-Tek, TGPH-090), upon which was spread a thin layer of uncatalysed (ketjenblack EC-300J) 10 wt.% teflonised carbon black. The catalysed layer in anode, consisting of 60% Pt:Ru (1:1) dispersed on carbon (from E-Tek) and bound with 10 wt.% Nafion<sup>®</sup>, from a solution of 5 wt.% Nafion<sup>®</sup> dissolved in mixture of water and lower aliphatic alcohol's (Aldrich), was spread on this diffusion backing layer. The catalyst loading in the anode was  $2 \text{ mg cm}^{-2}$  metal loading.

The cathode was constructed using a similar method as for the anode, using a thin diffusion layer bound with 10 wt.% PTFE, and  $1 \text{ mg cm}^{-2}$  Pt/C (60%)(from E-Tek) with 10 wt.% Nafion<sup>®</sup> in the catalyst layer. The electrodes were placed either side of a pre-treated ion conducting membrane. This pre-treatment involved boiling the membrane for 1 h in 5 vol.%  $\text{H}_2\text{O}_2$  and  $1 \text{ mol dm}^{-3}$   $\text{H}_2\text{SO}_4$  before washing in boiling deion water for 2 h with regular change of water. The assembly was hot-pressed at  $100 \text{ kg cm}^{-2}$  for 3 min at  $135 \text{ }^\circ\text{C}$ . The resulting MEA was installed in the cell after pressing, and hydrated with water circulated over the anode at  $75 \text{ }^\circ\text{C}$  for 48 h.

The membranes used were Nafion<sup>®</sup> 117, and radiation grafted polymer membranes, based on ethyleneterafluoroethylene (ETFE, from Du Pont) and polyvinylidene fluoride (PVDF, from Nowofol) reacted with styrene using the PIG technique [18]. Grafted ETFE-based polymer and PVDF-based polymer membrane were made with different thicknesses and different degrees of graft. The four grafted membranes used were: PVDF-g-PSSA membrane with 36% graft degree; PVDF-g-PSSA membrane with 14% graft degree (both PVDF-g-PSSA membranes with thickness of  $77 \text{ }\mu\text{m}$ ); ETFE-g-PSSA membrane with thickness of  $68 \text{ }\mu\text{m}$ , and ETFE-g-PSSA membrane with thickness of  $150 \text{ }\mu\text{m}$  (both ETFE-g-PSSA membranes with 27% graft degree).

Cell voltage versus current density response was measured galvanostatically, by incrementally increasing the current from open circuit and measuring the cell potential. Data are reported using the following standard conditions unless otherwise stated:

- methanol concentration:  $2 \text{ mol dm}^{-3}$  (M);
- methanol solution flow rate:  $5.9 \text{ cm}^3 \text{ min}^{-1}$ ;

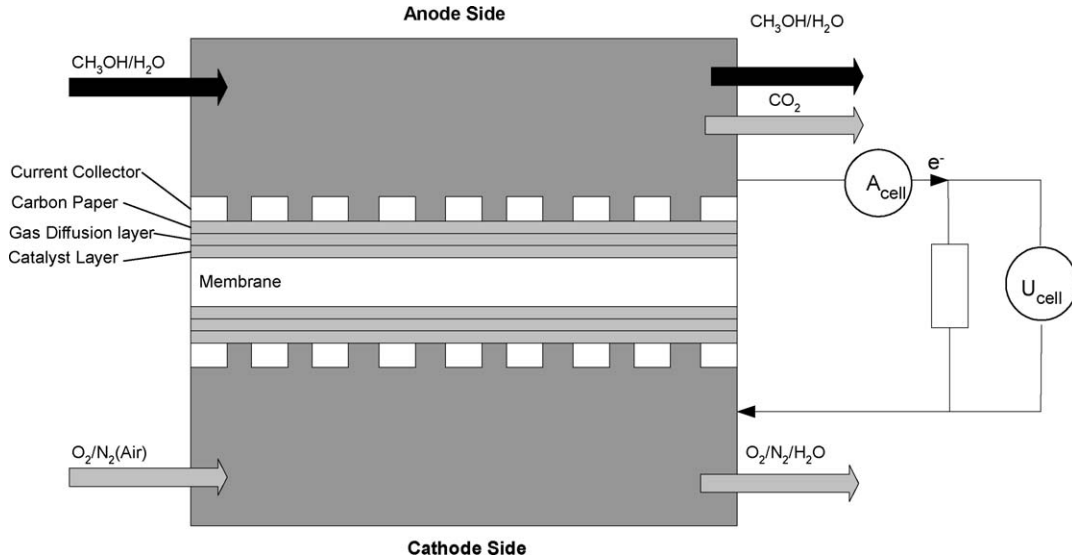


Fig. 1. Schematic drawing of DMFC.

- cell temperature: 80 °C;
- air flow rate: 570 cm<sup>3</sup> min<sup>-1</sup>;
- air pressure: ambient pressure.

### 3. Model formulation

#### 3.1. General regulation of power conversion

During the process of chemical energy transfer to electrical energy, the free energy of chemical reaction decides the equilibrium potential. Shen and Scott first raised the concept of power conversion coefficient and used it in modelling DMFC performance [19]. In their present work [17], they modified that the total electrical power produced by the DMFC is proportional to the square of the potential difference between the equilibrium potential and work potential. The modelling results with modified assumption were found to be close to experimental data, especially in the high operating current range. This approach is not only suited to DMFCs but also to other electrochemical power sources [17].

Here, if the equilibrium potential of a chemical power source is  $E_0$ , the work potential is  $E$ , which is defined in equation (5), and the total electric power produced with this chemical power source is  $P_{\text{total}}$ , the general regulation of power generated with chemical power sources can be expressed as below:

$$P_{\text{total}} = K(E_0 - E)^2 \quad (2)$$

where  $K$  is the power conversion coefficient, which decides the capacity of the chemical power source.

For a power source, we use  $I$  as the operating current or operating current density if considering power source of unit area,  $V$  is the export voltage,  $r$  is internal resistance and  $P$  is

the export electric power, then:

$$P = IV \quad (3)$$

$$P_{\text{total}} = IV + I^2r \quad (4)$$

$$E = V + Ir \quad (5)$$

From (2), (4) and (5) we obtain:

$$K(E_0 - V - Ir)^2 = IV + I^2r \quad (6)$$

From (6), the equation for the performance curve of a chemical power source without power loss can be written as:

$$V = E_0 - Ir - \frac{\sqrt{I^2 + 4KI E_0 - I}}{2K} \quad (7)$$

#### 3.2. The leak resistance and its effect in chemical power sources

In a practical power source, there is a leak resistance between the electrodes. Fig. 2 shows the electric circuit with leak resistance. If the value of the leak resistance is  $R$ , the leakage current is  $I_{\text{leak}}$ , the total current exported from power source is  $I_{\text{total}}$ , and the work current is  $I$ , then:

$$I_{\text{total}} = I + I_{\text{leak}} \quad (8)$$

$$I_{\text{leak}} = \frac{V}{R} \quad (9)$$

Considering  $I_{\text{leak}}$ , equation (6) can be written as:

$$K[E_0 - I_{\text{total}}r - V]^2 = I_{\text{total}}V + I_{\text{total}}^2r \quad (10)$$

Substituting (8) and (9) in equation (10) gives:

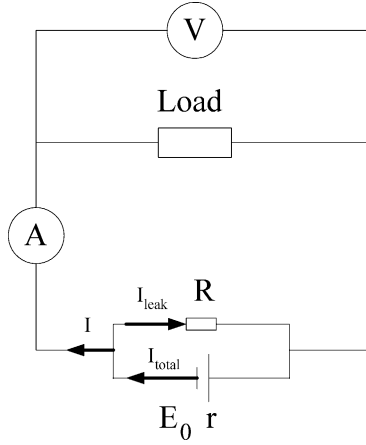


Fig. 2. The electric circuit with leak resistance.

$$\begin{aligned} & \frac{R+r}{R} K \left[ \frac{R}{R+r} E_0 - I \frac{Rr}{R+r} - V \right]^2 \\ &= \frac{V^2}{R} + IV \left( \frac{R+2r}{R+r} \right) + I^2 \frac{Rr}{R+r} \end{aligned} \quad (11)$$

Defining the term:

$$\begin{aligned} K' &= \frac{R+r}{R} K; & r' &= \frac{Rr}{R+r}; \\ E'_0 &= \frac{R}{R+r} E_0; & \beta &= \frac{R+2r}{R+r} \end{aligned} \quad (12)$$

gives:

$$\begin{aligned} \left( K' - \frac{1}{R} \right) V^2 - [2K'(E'_0 - Ir') + \beta I] V \\ + K'(E'_0 - Ir')^2 - I^2 r' = 0 \end{aligned} \quad (13)$$

When  $R \gg r'$ , and  $K' - \frac{1}{R} \approx K'$ ;  $\beta \approx 1$  (leak resistance is much greater than internal resistance) then:

$$V = E'_0 - Ir' - \frac{\sqrt{I^2 + 4K'(IE'_0 - I)}}{2K'} \quad (14)$$

For any value of internal resistance and leak resistance, the solution of equation (11) is:

$$V = R \frac{2K'(E'_0 - Ir') + \beta I - \sqrt{\Delta}}{2(K'R - 1)} \quad (15)$$

where

$$\begin{aligned} \Delta &= \left[ \beta^2 + 4K'r'(1 - \beta) + \frac{4r'}{R} (K'r' - 1) \right] I^2 \\ &+ 4K'E'_0 \left( \beta - \frac{2r'}{R} \right) I + \frac{4K'}{R} E_0'^2 \end{aligned} \quad (16)$$

When  $I=0$ , equation (15) can be written as:

$$V = \frac{E_0 R}{R+r + \sqrt{\frac{R+r}{K}}} \quad (17)$$

When  $V=0$ , we can obtain:

$$I_{\max} = \frac{E_0}{r + \sqrt{\frac{r}{K}}} \quad (18)$$

In practical operation, performance curves of chemical power sources are expressed as the plot of exported voltage,  $V$ , versus operating current,  $I$ . When considering the leak resistance, the performance curve can be expressed with equation (15). Open circuit potential (OCP) is expressed with equation (17).

### 3.3. Crossover and its effect in chemical power sources

Mass diffusion between anode and cathode will reduce the open circuit potential and the performance of chemical power sources. This is called crossover, which is a major problem in direct methanol and similar fuel cell. In chemical power sources, the crossover not only causes the corresponding parasitic electronic current [20], it also consumes the power converted from chemical energy to electrical energy.

If  $P_{\text{leak}}$  represents the power loss caused by mass diffusion between anode and cathode, then the total power exported by chemical power sources can be written as:

$$K(E_0 - Ir - V)^2 = IV + I^2 r + P_{\text{leak}} \quad (19)$$

$$\begin{aligned} KV^2 - [2K(E_0 - Ir) + I]V + K(E_0 - Ir)^2 - I^2 r \\ - P_{\text{leak}} = 0 \end{aligned} \quad (20)$$

The performance curve of a chemical power source with crossover is:

$$V = E_0 - Ir - \frac{\sqrt{I^2 + 4K(IE_0 + P_{\text{leak}}) - I}}{2K} \quad (21)$$

If  $I=0$ , the OCP is  $V_0$ :

$$K(E_0 - V_0)^2 = P_{\text{leak}} \quad (22)$$

$$V_0 = E_0 - \sqrt{\frac{P_{\text{leak}}}{K}} \quad (23)$$

## 4. Numerical results

In the ideal situation when omitting the power loss, the performance curve of a chemical power source can be expressed by equation (7). Figs. 3 and 4 show the typical numerical results of equation (7) with different internal resistance and different power conversion coefficient. As expected, the performance of the power source increases with increasing values of  $K$ . When the power conversion coefficient  $K \rightarrow \infty$ , we define the power source as a physical power source [17].

Fig. 5 shows typical numerical results of equation (15) with different values of leak resistance. As the value of leak resistance decreases, the open circuit potential and the performance of fuel cell decreases. Different performance curves converge at one point. When the value of leak resistance is higher than  $50 \Omega$ , which is 100 times the internal resistance,

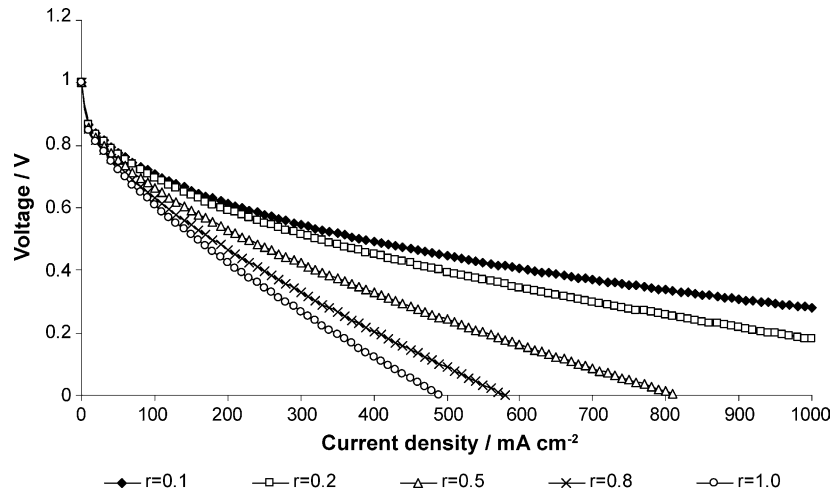


Fig. 3. Plot of equation (7) with different values of internal resistance. When  $E_0 = 1.0 \text{ V}$ ,  $K = 1.0 \Omega^{-1}$ , the unit of the inner resistance is  $\Omega \text{ cm}^{-2}$ .

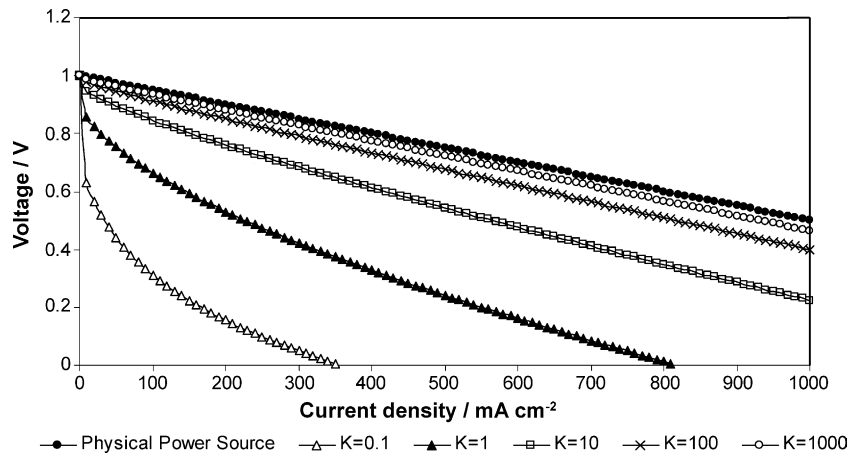


Fig. 4. Plot of equation (7) with different values of power conversion coefficient. When  $r = 0.5 \Omega \text{ cm}^{-2}$ ,  $E = 1.0 \text{ V}$ , the unit of the  $K$  is  $\Omega^{-1}$ .

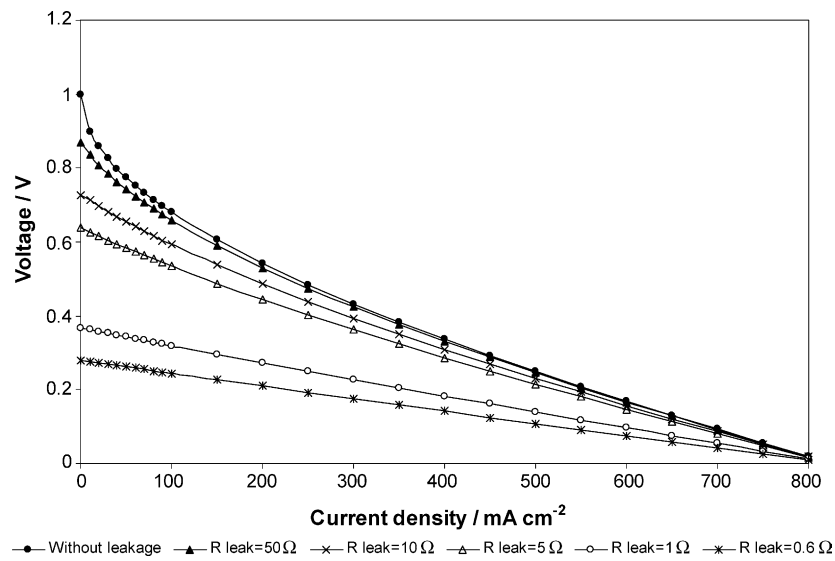


Fig. 5. Plot of equation (14) with different values of leak resistance. Here,  $r = 0.5 \Omega \text{ cm}^{-2}$ ,  $E = 1.0 \text{ V}$ ,  $K = 1.0 \Omega^{-1}$ .

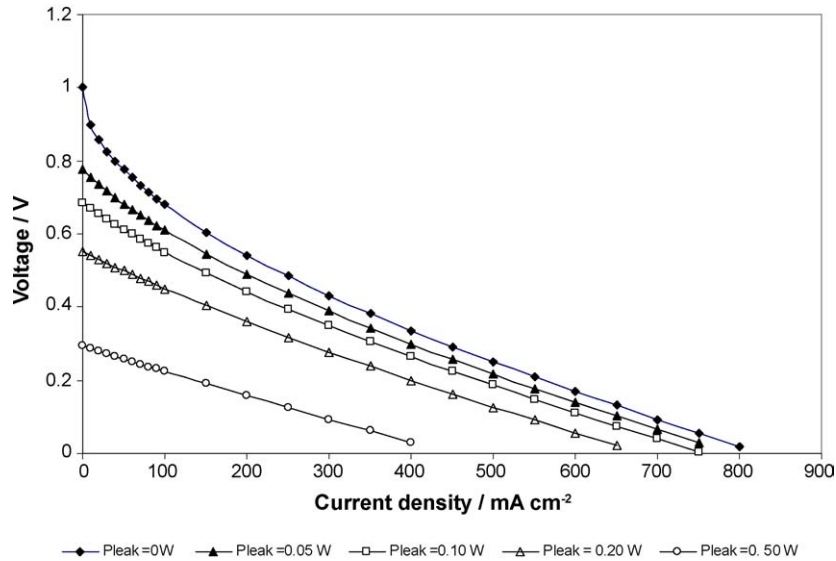


Fig. 6. Plot of equation (20) with different values of power loss. Here,  $r = 0.5 \Omega \text{ cm}^{-2}$ ,  $E = 1.0 \text{ V}$ ,  $K = 1.0 \Omega^{-1}$ .

the performance curve is close to the performance of a fuel cell without power loss.

Fig. 6 shows the numerical results of equation (21) with different values of power loss  $P_{\text{leak}}$ . As the value of  $P_{\text{leak}}$  increases, the open circuit potential and the performance of fuel cell decrease. The highest operating current density that can be achieved also decreases, so different performance curves do not converge at one point.

## 5. Analysis of direct methanol fuel cells

Fig. 7 shows DMFC performance with different ion conducting membranes. Fig. 7, and other researcher's [2,10,13,14] work, shows that the main cause of power loss in the DMFC is due to crossover. Barragán and Heinzl [12] has pointed out the relationship between methanol diffusion and open circuit potential of direct methanol fuel cells. Dohle et al. reported [10] that the methanol diffusion flux decreased as the operating current increases. Here, we apply equation (21) to the performance of the DMFCs. Although  $P_{\text{leak}}$  varies with operating current, to simplify the situation, we assume  $P_{\text{leak}}$  is a constant. From Fig. 6, we see that the value of  $P_{\text{leak}}$  plays a more important rule at low operating current.

From the DMFC fuel cell performance curves in Fig. 7, we can calculate fuel cell parameters:  $K$ ,  $r$ , and  $P_{\text{leak}}$  in equa-

tion (21). The nonlinear least square regression problem is solved by using the Gauss–Newton method implemented in MATLAB. Table 1 shows results of the different DMFCs parameters:  $K$ ,  $r$ , and  $P_{\text{leak}}$  obtained.

The value  $K$  reflects the chemical characteristics of electrodes. The higher the value of  $K$  means a higher rate of energy conversion. A higher value of internal resistance ( $r$ ) means more electrical power will be consumed in the internal resistance and the fuel cell has lower performance. The higher the value of  $P_{\text{leak}}$  means that a greater electrical power is consumed by crossover and then the performance of fuel cell will be lower.

Considering the power loss of the DMFC caused by methanol crossover, the value of  $P_{\text{leak}}$  is related to methanol mass diffusion flux, by:

$$P_{\text{leak}} = k \Delta G J_{\text{MeOH}} \quad (24)$$

where  $J_{\text{MeOH}}$  is the methanol crossover flux;  $\Delta G$  is the free energy of methanol oxidation and  $k$  is a proportionality coefficient.

If every methanol molecule that diffuses to the cathode reacts with oxygen immediately, then the proportionality coefficient  $k$  is equal to 1. Then, equation (24) can be written as:

$$P_{\text{leak}} = \Delta G J_{\text{MeOH}} \quad (25)$$

Table 1  
Fuel cell parameters of different DMFCs

Fuel cell membrane	PVDF-g-PSSA graft (36%)	PVDF-g-PSSA graft (14%)	ETFE-g-PSSA (68 $\mu\text{m}$ )	ETFE-g-PSSA (150 $\mu\text{m}$ )	Nafion <sup>®</sup> 117
$E_0$ (V)	1.21	1.21	1.21	1.21	1.21
$K$ ( $\Omega^{-1}$ )	0.126	0.0434	0.081	0.046	0.184
$r$ ( $\Omega \text{ cm}^{-2}$ )	0.044	0.6	0.092	1.16	0.698
$P_{\text{leak}}$ (W)	0.0467	0.0205	0.0391	0.0234	0.0778
$J_{\text{MeOH}}$ ( $10^{-6} \text{ mol min}^{-1} \text{ cm}^2$ )	4	1.8	3.4	2	6.7



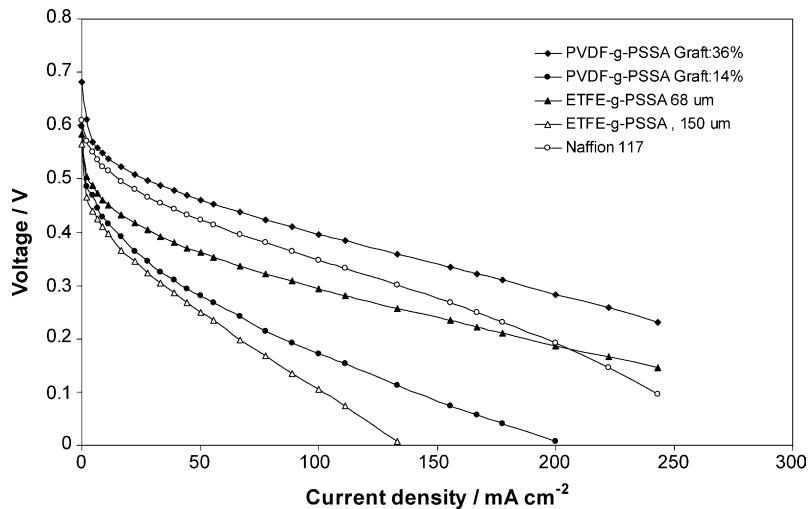


Fig. 7. Performance curve of DMFC with different membranes.

With equation (25), we can calculate the value of  $J_{\text{MeOH}}$  with the fuel cell constant  $P_{\text{leak}}$ . Table 1 shows the calculated values of methanol crossover of the fuel cell. Comparing the calculated value of methanol crossover with experimental data by other scientists, we find our data is much smaller than the methanol crossover in Nafion<sup>®</sup> 115, i.e. approximately  $5 \times 10^{-6} \text{ mol cm}^2 \text{ s}^{-1}$  [9], but it is close to the data supplied by Arico et al. [21] which is around  $2\text{--}5 \times 10^{-6} \text{ mol cm}^2 \text{ min}^{-1}$  for grafted ETFE-based polymer membranes.

In Table 1, we compare the performance of five direct methanol fuel cells with different membranes. The fuel cell made with Nafion<sup>®</sup> 117 has the highest power conversion constants, but the crossover constant is also the highest; the internal resistance is also quite high. The fuel cell with a grafted PVDF membrane has the lowest crossover constant

and internal resistance. So, its performance is highest. Decreasing the degree of grafting will decrease the crossover constant, but the internal resistance will also increase. The optimum graft degree is 36%. The fuel cell with a grafted ETFE based polymer membrane has a relatively lower internal resistance and lower crossover constant. Although an increase in the thickness of the membrane will decrease the crossover rate, the internal resistance of the fuel cell will increase as well. The membrane of thickness  $68 \mu\text{m}$  has a good performance.

Table 1 shows the fuel cell parameters of different DMFCs. Applying these fuel cell parameters in equation (21), we can calculate values of the fuel cell performance. Fig. 8 shows the modelling results of performance curves of these DMFCs. As shown in Figs. 8 and 7, we find the modelling result agrees with the performance curves of practical DMFCs.

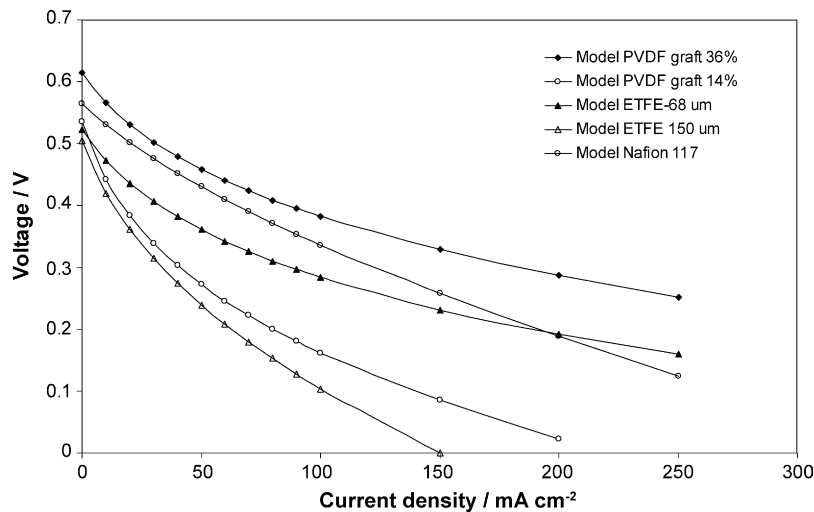


Fig. 8. The calculating value of performance curve with different membranes.

## 6. Conclusion

Leak resistance and crossover in fuel cells can cause power loss in fuel cell performance. With a general regulation of power converted from chemical energy to electrical energy, the fuel cell performances with two kinds of power losses can be expressed mathematically. For direct methanol fuel cells, methanol crossover is the main reason for the power loss. The mathematical expression of the performance of fuel cell with the crossover and fuel cell parameters can elucidate the DMFC characteristics. The mathematical model of performance gives good prediction of the practical performance.

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