

The effect of the MEA preparation procedure on both ethanol crossover and DEFC performance

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Abstract

In the present work, the changes of Nafion[®]-115 membrane porosity in the presence of ethanol aqueous solutions of different concentrations were determined by weighing vacuum-dried and ethanol solution-equilibrated membranes. It was found that membrane porosity increases as ethanol concentration increases. Membrane electrode assemblies (MEAs) have been prepared by following both the conventional and the decal transfer method. The ethanol crossover through these two MEAs was electrochemically quantified by a voltammetric method. A 10 h stability test of direct ethanol fuel cell (DEFC) at a current density of 50 mA cm⁻² was carried out. It was found that the electrode preparation procedure has an obvious effect on ethanol crossover and direct ethanol fuel cell's performance and stability. The single DEFC test results showed that about 15 and 34% of the original peak power density was lost after 10 h of life test for the MEAs prepared by the decal transfer method and the conventional method, respectively. Electrochemical impedance spectrum (EIS) results of the MEAs showed that, in the case of the membrane electrode assembly prepared by the following decal transfer method, the internal cell resistance was almost the same, 0.236 Ω cm² before the life test and 0.239 Ω cm² after 10 h of life test, while the respective values for the membrane electrode assembly by the conventional method are 0.289 and 0.435 Ω cm². It is supposed that the improved cell performance with MEA by the decal transfer method could be resorted to both a better contact between the catalyst layer and the electrolyte membrane and higher catalyst utilization. Furthermore, based on the experimental results, the increased internal cell resistance and the degraded single DEFC performance could be attributed to the delamination of the catalyst layer from the electrolyte membrane.

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1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) have been actively and widely developed for both stationary and mobile applications due to their compactness, zero-pollution, high output energy density at room temperature and their excellent easiness of start-up and shut-down in the system

operation [1–5]. However, there are difficulties in hydrogen supply infrastructure or fuel reforming technology with the clean-up of impurities such as carbon monoxide and sulfur compounds. Undoubtedly, one of the most direct solutions to the fueling problem would be to develop fuel cells that operate on a liquid fuel. Direct methanol fuel cells (DMFCs) had been actively investigated since 1960s [6] and much progress has been made in recent years [7]. However, the perfluorosulphonate, Nafion[®] membranes manufactured by E.I. Du Pont Nemours and Co. in 1972 are quite permeable for methanol [8,9] to the oxygen cathode, which is one of the main factors significantly affecting DMFCs performance, and in this way lowering the fuel cell efficiency with a loss

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of more than 50% at lower current densities [10]. In addition, methanol is relatively toxic and inflammable, and it is neither a primary nor a renewable fuel. On the other hand, as a naturally available and renewable material, ethanol is considered to be an attractive substitute for methanol with positive impact on both economy and environment [11–13]. Moreover, ethanol combustion will cause no rise in global concentrations of greenhouse gases. It should also be noted that ethanol itself and the intermediate products of its oxidation are less toxic than the other alcohols [14]. To our knowledge, the research and development of DEFC have been concentrated on both the ethanol electro-oxidation mechanism [15–19] and the identification of ethanol oxidation products over electrocatalysts [20–23]. There are few publications concerning the performance [24] of DEFC and ethanol crossover from the anode to the cathode through Nafion[®] membrane [25,26].

In the present investigation, Nafion[®] membrane porosity in the presence of the ethanol aqueous solutions with different concentrations at ambient temperature was determined. Membrane electrode assemblies were prepared by both the conventional method and the decal transfer method. The ethanol crossover has been electrochemically determined by the voltammetric method. A 10 h DEFC life test at a current density of 50 mA cm^{-2} was carried out. The techniques of single fuel cell test, electrochemical impedance spectroscopy (EIS) were applied to investigate the cell performance degradation of the MEAs fabricated by different methods: (a) the conventional method and (b) the decal transfer method.

2. Experimental

2.1. Membrane porosity measurement

Nafion[®]-115 membranes were adequately pretreated by boiling them successively in 3 wt.% hydrogen peroxide, deionized water, 0.5 mol L^{-1} sulfuric acid and deionized water again, 1 h for each step [27,28]. In order to quantify membrane porosity, the Nafion[®]-115 membrane samples were immersed in ethanol aqueous solutions with different ethanol concentrations and equilibrated for 36 h at ambient temperature. After that, the membrane was removed from the solution and the excess liquid was wiped from its surface by the use of a filter paper. The weight of swollen membranes was determined by using an accurate balance. After drying at 60°C in vacuum for 24 h the weight of the membranes was determined again. Each sample porosity was determined for at least three times and the results were within the experimental error.

2.2. Membrane electrode assembly (MEA) preparation

MEAs were fabricated by the conventional method (CON) and the modified decal transfer method (DTM) for electrode preparation which both are schematically represented in Fig. 1. For the sake of simplicity, the MEAs prepared by

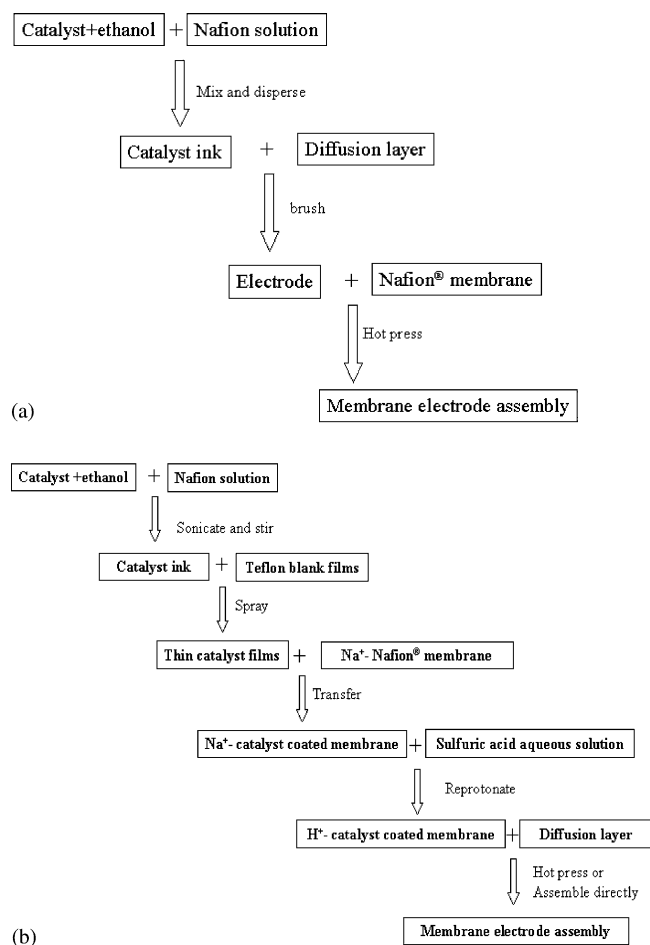


Fig. 1. Schematic presentation of the detailed electrode preparation procedures for: (a) the conventional method and (b) the decal transfer method.

the CON or the DTM are here denoted as MEA-CON and MEA-DTM, respectively. The main difference between these two electrode preparation procedures is that the catalyst layer was applied onto the gas diffusion layers for MEA-CON or onto the electrolyte membrane for MEA-DTM with a transfer process at higher temperature value. The conventional electrode preparation procedure has been previously described in detail [29]. Here, a detailed procedure for the DTM is presented. Pre-treated H⁺ form Nafion[®]-115 membranes were converted into the Na⁺ form by successively boiling them in 0.5 mol L^{-1} NaOH aqueous solution and deionized water, for 1 h each step. PtRu black of nominal 1:1 atomic ratio and Pt black purchased from Johnson Matthey Corp. were used as the anode and cathode catalysts, respectively. Catalyst inks were prepared by adding 5% of Nafion solution (1100 equiv. weight) to the pre-wetted by water and ethanol well-dispersed catalyst inks. The solubilized Nafion was converted into the Na⁺ form by adding an appropriate amount of NaOH aqueous solution. Appropriate amounts of well-dispersed anode and cathode inks were uniformly sprayed by a gunnite to Teflon decal blanks with a given size to give metal loadings of about 3.0 mg cm^{-2} for both the anode and the cathode, respectively. The *catalyst-coated membrane* (CCM)

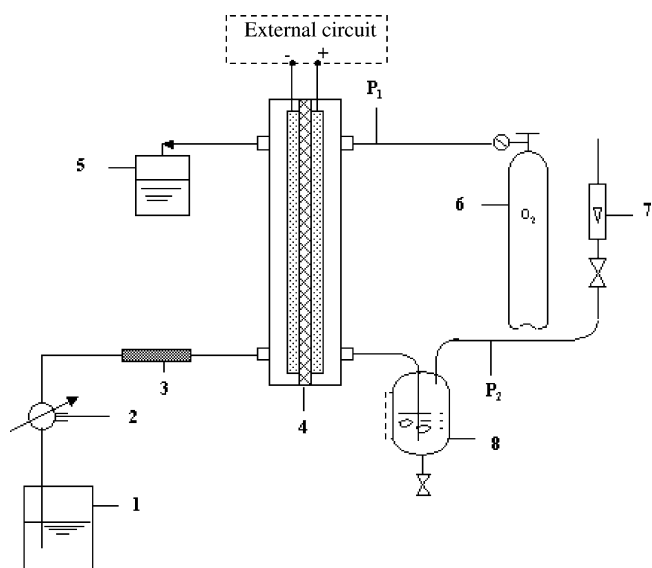
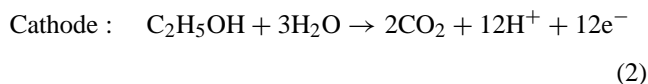
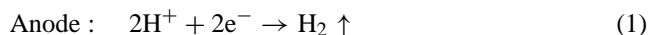


Fig. 2. Schematic configuration of a single DEFC test apparatus: (1) ethanol aqueous solution tank; (2) pump; (3) heater; (4) fuel cell; (5) ethanol aqueous solution recovery tank; (6) oxygen cylinder; (7) flow meter; (8) separator.

was obtained by transferring the catalyst films from the Teflon films to the Na⁺-Nafion[®]-115 membrane by preheating at 160–180 °C for 60 s to remove water from the membranes and by hot pressing at 160–180 °C and 100 kg cm⁻² for another 90 s. The CCM was adequately reprotinated in 0.5 mol L⁻¹ H₂SO₄ for 2 h and then immersed in deionized water for 1 h in a 80 °C water bath. The CCM was sandwiched between two 2 cm × 2 cm diffusion layers and then this assembly was inserted into the fuel cell hardware described previously [30]. Here, it is worth noting that the MEA obtained by the decal transfer method can present a better cell performance when it is used in direct methanol fuel cell systems [30].

2.3. Ethanol crossover measurements by a voltammetric method

Ethanol crossover measurements were carried out by using an EG&G potentiostat/galvanostat 273A and in-house made fuel cell test apparatus presented in Fig. 2. The ethanol aqueous solution was pumped to the anode compartment of the fuel cell without backpressure and high purity nitrogen was supplied at 2 atm to the cathode. The reactions taking place at the anode and the cathode are described as follows:



From the above two equations, it is suggested that hydrogen evolution reaction takes place at the anode of the fuel cell and the permeated ethanol from the anode to the cathode is oxidized at the cathode of the fuel cell, respectively. The anode can act not only as the counter electrode but also as

the *dynamic hydrogen reference electrode* (DHE) [31,32]. By applying a dynamic potential versus DHE, to pump hydrogen ions to the anode of the fuel cell, the ethanol crossover measurements were electrochemically performed at different temperatures to investigate the influence of the MEA preparation procedure on the ethanol crossover through MEAs. The potential range was from 0 to 1.2 V versus DHE and the potential scanning rate was 2.0 mV s⁻¹.

2.4. Single fuel cell test

The single fuel cell tests in the present investigation were carried out in the test apparatus shown in Fig. 2. Prior to data collection, the MEA was fully hydrated by pumping deionized water to the anode side at 75 °C for about 4 h. During the operation of DEFC, a diluted ethanol aqueous solution preheated at the same temperature as the cell working temperature, was fed into the inlet located at the bottom of the anode at a flow rate of 1.0 mL min⁻¹ by a peristaltic pump without back pressure. At the same time, non-humidified ambient temperature oxygen was supplied to the inlet located at the top of the cathode side at 2 atm controlled by a pressure regulator. The polarization curves were obtained using a Fuel Cell Test System (Arbin Instrument Corporation) in a galvanodynamic polarization mode.

2.5. AC-impedance characterization

AC-impedance spectra of the DEFC were collected under open circuit conditions by using a Lock-in Amplifier (EG&G Model 5210) coupled with a potentiostat/galvanostat (EG&G Model 273A) in a frequency range between 6000 and 0.1 Hz with 10 points per decade. The amplitude of the AC-voltage was 5 mV.

3. Results and discussion

Membrane porosity and membrane water content will contribute to the overall transport characteristics of an ion exchange membrane [33] and thus play an important role in the PEMFC performance. Wet membrane porosity (ϵ) was determined at ambient temperature by weighing vacuum-dried and ethanol aqueous solutions equilibrated membranes. The membrane porosity for Nafion[®]-115 was calculated by using Eq. (3) [34,35]:

$$\epsilon = \frac{\text{fluid uptake volume}}{\text{total volume}} = \frac{(W_{\text{wet}} - W_{\text{dry}})\rho_{\text{dry}}}{(W_{\text{wet}} - W_{\text{dry}})\rho_{\text{dry}} + W_{\text{dry}}\rho_{\text{sol}}} \quad (3)$$

where ρ_{dry} is the dry membrane density (2.075 g cm⁻³, for dry proton-form Nafion[®] membranes), ρ_{sol} the ethanol aqueous solution density and W_{wet} and W_{dry} the wet membrane weight and the dry membrane weight, respectively.

It can be clearly seen from Table 1 that the membrane porosity increases as the ethanol concentration increases. It

Table 1
Nafion®-115 membrane porosity in ethanol aqueous solutions with different concentrations at ambient temperature

C_{ethanol} (mol L ⁻¹)	Membrane porosity ε
0.5	0.3706
1.0	0.3862
2.0	0.4162
4.0	0.5012
8.0	0.6433

is known that the change in the membrane porosity due to ethanol uptake, which leads to the swelling of the membrane, is a reversible process. The membrane porosity increase means that the membrane swells to a higher degree as the ethanol concentration increases. This phenomenon can be explained by taking into account the electrostatic interactions in the polymer. According to Hsu and Gierke [36], Nafion® membrane is described as a series of clusters interconnected by narrow pores. In each cluster, the fixed membrane charges must create an electrostatic field, which extends inward the center of each sphere. Within this region, the size of the ionic atmosphere can thus be described by the Debye length defined as

$$\lambda = \left[\frac{\varepsilon RT}{2F^2 \bar{c}} \right]^{1/2} \quad (4)$$

where ε is the relative permittivity of water, F the Faraday constant and \bar{c} the ion concentration in the cluster. On the other hand, it is known that the polarity of ethanol is less than that of water. Therefore when the aqueous ethanol concentration is increased, \bar{c} becomes smaller, and thus the size of the ionic atmosphere becomes larger and it can be assumed that the cluster size is increased as well. Thus, Nafion® membrane will present a higher swelling degree in ethanol solutions with higher concentrations. Considering the delamination of the catalyst layer from the membrane, it should be noted that when ethanol is served as the fuel for direct alcohol fuel cells, the swelling of Nafion® membrane in ethanol solutions will lead to a distortion difference between the electrode and the electrolyte, resulting in the readily delaminating of the catalyst layer from the membrane. Consequently, this will deteriorate the cell performance and shorten the longevity of direct ethanol fuel cells.

Fig. 3 shows the ethanol crossover current density versus applied voltage in the MEA-CON and MEA-DTM at different temperature values, respectively, with 1.0 mol L⁻¹ ethanol pumped to the anode. It is assumed that all the permeated ethanol from the anode to the cathode was electrochemically converted in the catalyst layer of the cathode. The more the permeated ethanol is, the higher the current density corresponding to the ethanol electro-oxidation will be. In Fig. 3 it can be distinguished that the current density increases as the temperature increases. In other words, this means that ethanol crossover increases with the temperature increment [26]. It can also be clearly seen from Fig. 3 that in both cases there is a peak current density as the applied voltage is in-

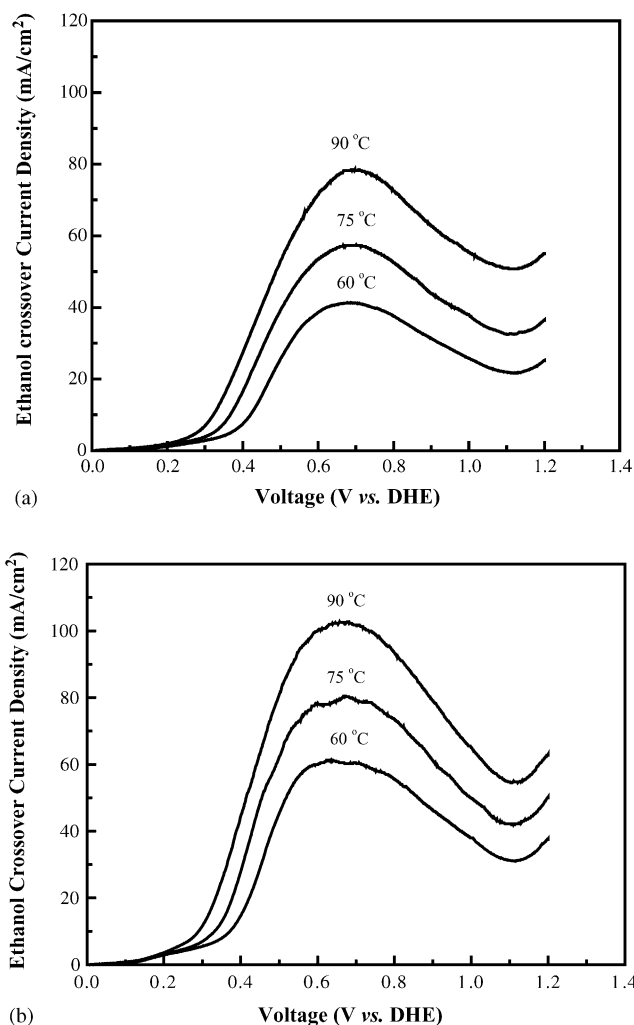


Fig. 3. Comparison of ethanol crossover rate through the MEAs by using different electrode preparation methods at different temperatures: (a) the conventional method; (b) the decal transfer method. Anode: PtRu black (Pt:Ru = 1:1, Johnson Matthey Corp.), 3.0 mg (Pt + Ru) cm⁻², $C_{\text{ethanol}} = 1.0 \text{ mol L}^{-1}$, flow rate: 1.0 mL min⁻¹. Cathode: Pt black (Johnson Matthey Corp.), 3.0 mg Pt cm⁻², $P_{\text{N}_2} = 2.0 \text{ atm}$. Electrolyte: Nafion®-115 membrane.

creased and it is worth noticing that the observed behavior does not occur during methanol and formic acid crossover, in which there is always a transport-controlled limiting current density at the Pt/membrane interface of the cathode side [31,32,37]. In the present case, the observed peak current density might be attributed to the higher activation over-potential of ethanol electro-oxidation over Pt catalysts compared to methanol or formic acid electro-oxidation. However, under the same operation conditions, the current density for the permeated ethanol electro-oxidation at a given potential (versus DHE) could also be used for ethanol crossover quantification. The potential of 0.5 V versus DHE was selected as the comparative point and the results are summarized in Table 2. One can distinguish jointly from both Fig. 3 and Table 2 that the MEA-DTM shows a higher ethanol crossover current density in comparison with the MEA-CON. The former is almost 1.5 times higher than the latter at almost all the investigated tem-

Table 2

Comparison of the current density for ethanol crossover through both MEA-CON and MEA-DTM by the voltammetry technique at different temperatures

	Ethanol crossover current density (mA cm^{-2}) at 0.5 V vs. DHE	
	MEA-CON	MEA-DTM
60 °C	25	45
75 °C	39	62
90 °C	53	80

Table 3

Comparison of ethanol crossover apparent activation energies through both MEA-CON and MEA-DTM determined by the voltammetry technique and through pure Nafion[®] membrane by the GC method

	Bare Nafion [®] membrane	MEA-CON	MEA-DTM
Activation energy (kJ mol^{-1})	24.9	25.2	19.3

peratures. The higher ethanol crossover in the MEA-DTM could be caused by the thinner membrane which results from the high temperature transfer process involved in the decal transfer electrode preparation method shown in Fig. 1(b). On the other hand, the apparent activation energies for ethanol crossover through MEA-CON or MEA-DTM by the voltammetric method technique and through bare Nafion[®] membrane by the GC method, respectively, were also compared and shown in Table 3. It can be clearly seen from Table 3 that in the cases of bare Nafion[®] membrane by the GC method and MEA-CON by the voltammetry technique, the apparent activation energies for ethanol crossover shows almost the same values, 24.9 kJ mol^{-1} for the former and 25.2 kJ mol^{-1} for the latter, respectively. The slightly higher activation energy value for the MEA-CON could be attributed to the effect of electro-osmotic drag when the voltammetry technique was used. However, in the case of the MEA-DTM by the voltammetry technique, a smaller apparent activation energy value (19.3 kJ mol^{-1}) was calculated. This may be attributed to the thinner membrane that resulted from the high temperature transfer process involved in the decal transfer method shown above in Fig. 1(b).

A 10 h life test of the single DEFC at 90 °C at a current density of 50 mA cm^{-2} is shown in Fig. 4. It can be clearly seen from Fig. 4 that there are some spikes on the cell voltage versus time figure, and this was caused by the interrupt of the continuous discharge of DEFC due to the drainage at the cathode during the process of life test. These spikes might primarily be caused by the reduced ethanol crossover effect on the cathode performance and the decreased flooding of the cathode. From Fig. 4 it can also be seen that in the case of the MEA-DTM, there is a decay in the output voltage from initial 0.425 V to the final 0.312 V after 10 h discharge with a voltage attenuation of 26.6%, while the corresponding value for MEA-CON is from 0.434 to 0.283 V and its voltage attenuation is about 34.8%. From the same figure, it can also be observed that during the whole discharge process, the sin-

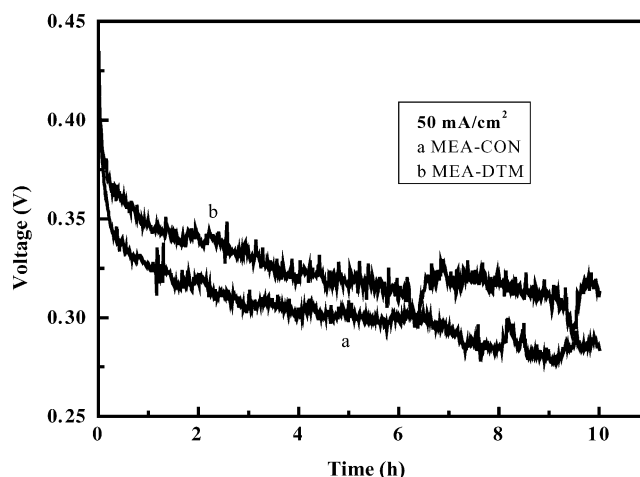


Fig. 4. Stability test data for the single DEFCs: (a) MEA-CON; (b) MEA-DTM. $T_{\text{cell}} = 90 \text{ }^\circ\text{C}$; anode: PtRu black (Pt:Ru = 1:1, Johnson Matthey Corp.), $3.0 \text{ mg (Pt + Ru) cm}^{-2}$, $C_{\text{ethanol}} = 1.0 \text{ mol L}^{-1}$, flow rate: 1.0 mL min^{-1} . Cathode: Pt black (Johnson Matthey Corp.), $3.0 \text{ mg Pt cm}^{-2}$, $P_{\text{O}_2} = 2.0 \text{ atm}$. Electrolyte: Nafion[®]-115 membrane.

gle DEFC with the MEA-DTM exhibits higher output cell voltage than that with the MEA-CON with an exception in the very initial time. This better cell performance could be attributed to a better contact between the catalyst layer and the membrane, while a little lower initial cell voltage might be related to the effect of ethanol crossover. This is due to the fact that the electric circuit is open before discharge and in this case the MEA-DTM presents a higher ethanol crossover because of the thinner membrane as shown in Fig. 3 and Table 2. On the other hand, Shukla et al. [38] reported that the MEA after a DMFC life test recovered its initial performance when the durability test was stopped. However, in the present case, the voltage loss cannot be fully recovered after the 10 h durability test, as it is shown by the following single fuel cell tests. This may suggest that there is an irreversible decline process in the DEFC life test procedure. Among all the possible factors affecting the performance stability of DEFC, the delamination of the catalyst layers from the electrolyte membrane might be one of the main irreversible ones leading to the unrecoverable cell performance decline during the process of life test. The delamination phenomena can be directly and obviously observed after having opened the investigated cell, especially for the MEA-CON.

Fig. 5 depicts the polarization curves before and after the life test at 90 °C with oxygen at 2 atm using Nafion[®]-115 membrane as the electrolyte. It can be observed from Fig. 5 that, the open circuit voltages are about 0.640 and 0.600 V for the DEFCs with MEA-CON and MEA-DTM, respectively. In the case of MEA-DTM, the lower DEFC's open circuit voltage could be caused by the higher ethanol crossover in the MEA-DTM shown above in Fig. 3 and Table 2. From Fig. 5, one can also distinguish that, in the case of MEA-DTM, the output cell voltages of the single DEFC at 100 mA cm^{-2} decline from initial 0.306 to 0.286 V and the respective values are 0.284 and 0.204 V for the MEA-CON after the 10 h

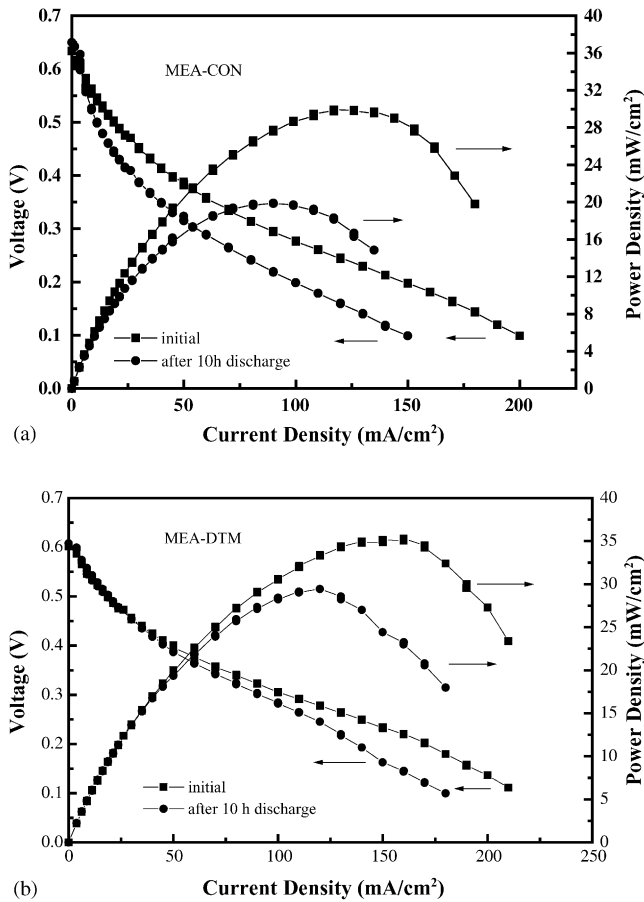


Fig. 5. Polarization and power density data for the single DEFCs before and after the life test under the same operating conditions as those in Fig. 3: (a) MEA-CON; (b) MEA-DTM.

life test. It can also be clearly seen from Fig. 5 that when the DEFC adopted the MEA-DTM, the peak power densities are 35.4 mW cm^{-2} at 160.0 mA cm^{-2} , and 30.0 mW cm^{-2} at 120 mA cm^{-2} for before and after the life test, respectively, with about 15% of the maximum power density lost after the life test. While in the MEA-CON case, the maximum power densities are 29.9 mW cm^{-2} at 140 mA cm^{-2} and 19.9 mW cm^{-2} at 100 mA cm^{-2} for before and after the 10 h life test, respectively, and the maximum power density loss during this discharge process is about 33.4%. By comparing these two cases, one could conclude that DEFC with the MEA-DTM presents obviously better performance than that with the MEA-CON. In general, a large area of electrochemical active interface between the catalyst layer and the membrane is usually desired for attaining maximum energy output by a fuel cell. From this point of view, the MEA-DTM has a higher electrochemical active interface than the MEA-CON. After the life test, the undesired cell performance might be due to the inadequate catalyst/membrane interface, which could be caused by the catalyst layer delamination from the membrane due to the different swelling degree between the catalyst layer and the electrolyte with ethanol aqueous solution supplied to the anode compartment. Moreover, the single

DEFC with MEA-CON shows a bigger slope in ohmic polarization region than that with MEA-DTM under the same operation conditions. Furthermore, it can also be seen from Fig. 5 that the cell voltage of the single DEFC after the life test falls more rapidly in the ohmic polarization region in comparison with that before the life test for both MEAs prepared by different electrode preparation methods. This is probably an indication of an increased internal resistance of the cell after the life test.

AC-impedance spectra were collected in order to determine the cell internal resistance before and after the 10 h life test. The resistance was determined by the high frequency intercept of the AC-impedance response on the real axis of the Nyquist plot. The Nyquist diagrams under open circuit conditions at 90°C before and after the life test are presented in Fig. 6. It can be clearly seen from Fig. 6 that when the MEA was prepared by the DTM, the typical resistance values under these operation conditions were 0.236 and $0.239 \Omega \text{ cm}^2$ before and after the 10 h life test at 90°C , while in the case

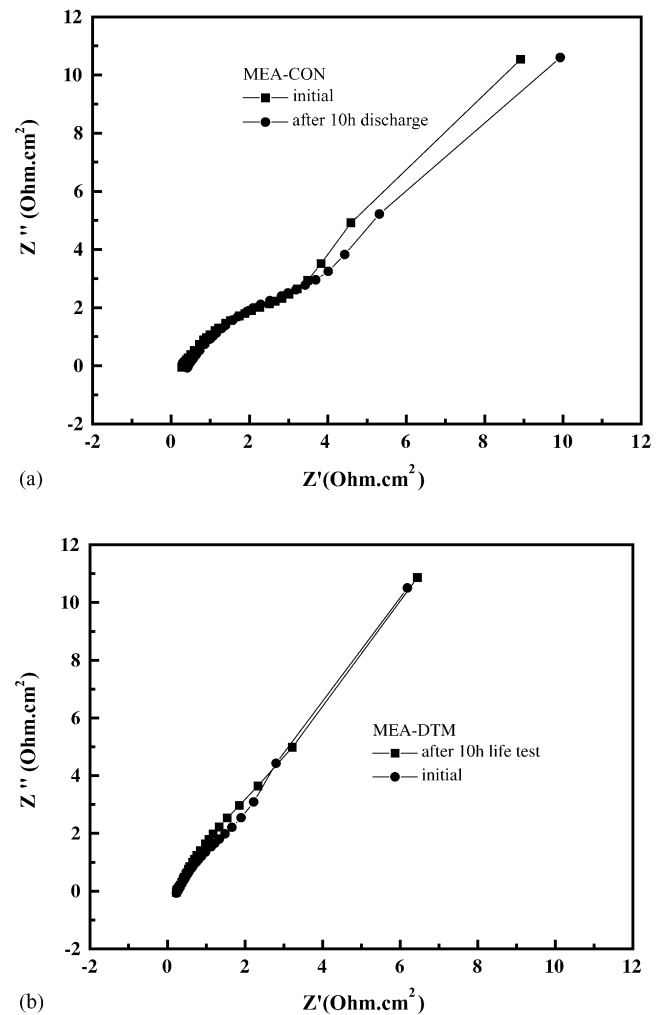


Fig. 6. Nyquist diagrams of the DEFC at open circuit before and after the life test under the same operation conditions as those described in Fig. 3: (a) MEA-CON; (b) MEA-DTM.

of the MEA-CON, the corresponding values are 0.289 and $0.435 \Omega \text{ cm}^2$, respectively. Based on these values, one can distinguish that, in the case of MEA-DTM, the internal cell resistance are almost the same for before and after the 10 h life test, while in the latter case the corresponding value is increased by 1.5 times after the 10 h life test. Such an obvious increase in internal resistance can lead to a significant decline in the performance of DEFC, which can also be observed in Fig. 5. Based on the above experimental results, for the case of MEA-CON, the obviously increased internal cell resistance after the life test with respect to that before the life test could be attributed to the difficulty in electron transporting and proton flowing at three phase interface, and this might be caused by the inadequate contact between the catalyst layer and the electrolyte resulting from the catalyst layer delamination from the electrolyte due to the different swelling degree of the electrode and the electrolyte membrane when ethanol aqueous solutions are continuously supplied to the anode compartment of the cell. At the same time, it can also be distinguished that in the case of the MEA-CON, the DEFC always presents higher internal cell resistance than that with the MEA-DTM under the same operation conditions, and the former has a more obvious increase in the internal cell resistance than the latter after the 10 h life test. The inferior cell performance with the MEA-CON could be attributed to the lower catalyst utilization and the undesired contact between the catalyst layer and the electrolyte. In other respect, the lower internal cell resistance of the MEA-DTM could resort to the thinner electrolyte membrane due to the transfer process during the electrode preparation procedure. Furthermore, compared to the DEFC with the MEA-DTM, the more obvious cell resistance increment of the one with the MEA-CON might be caused by the catalyst layer delamination from the electrolyte due to the fact that the relatively rigid catalyst matrix at the electrodes cannot easily accommodate the swelling of the electrolyte membrane in the liquid-DEFC.

4. Conclusion

Nafion[®] Membrane has an increased swelling degree with ethanol concentration increment. Membrane swelling can have a serious effect on electrode structure, leading to a readily delaminating of the catalyst layer from the electrolyte membrane when ethanol serves as the fuel for direct alcohol fuel cells. The MEA prepared by the decal transfer method presents higher ethanol crossover through Nafion[®] membrane because of the thinner membrane resulting from the high temperature transfer process involved in the electrode preparation procedure. About 15 and 34% of the initial peak power density of single DEFCs were lost after a 10 h life test in which the MEAs was prepared by the decal transfer method and the conventional method, respectively. The cell internal resistance increased significantly after the life test compared to the original cell internal resistance for the MEA-CON, while for the MEA-DTM case there is almost no change in

the cell resistance. This might be attributed to the catalyst layer delamination from the membrane due to the different degrees of swelling of the electrode and the membrane. It can also be found that the increase in ohmic losses contributes only to a small fraction to the loss of the cell performance, as the current densities are small. The single DEFC with the MEA-DTM exhibited improved performance and lower internal resistance in comparison with one with the MEA-CON, mainly due to a better contact between the catalyst layer and the electrolyte membrane and higher catalyst utilization. It should also be noted that there are still some other reasons for the degradation of DEFC such as the agglomeration of electrocatalysts, but in such a short life test, the catalyst layer delamination from the electrolyte membrane may be the key factor deteriorating the single DEFC performance and the stability.

Acknowledgements

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