THE MANUFACTURE ELECTRODE PROPERTIES ON THE HYDROGEN REACTION KINETIC IN PMFC

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Abstract

Economic and environmental issues are forcing industry to explore alternative energy sources for vehicles and electronic devices. Much effort is being put into fuel cell research because they hold the promise of high energy density, low environmental impact, and broad applicability. Hydrogen is the main source of fuel for high-performance applications with water being the only product. These attributes make the hydrogen fuel cell a practical replacement for many power sources that are inefficient and environmentally unfriendly. The basic functional unit of the fuel cell is the membrane electrode assembly (MEA). The MEA is the loci where the electrochemistry occurs to produce electricity from hydrogen and oxygen. The MEA consists of the polymer membrane with an electrode adhered to either side. The electrode contains the platinum electro-catalyst dispersed on carbon black with a polymer binder. The effect of gas diffusion electrode to the performance of fuel cells was studied in a single cell. Performance dependence on the intrinsic material properties was studied by Fuel cell test system. The interdependence between gas diffusion electrode and fuel cell performance was covered. Long-term stability of the PEMFC stack built in the Laboratory of Advanced Energy Systems was demonstrated by operating it continuously.

Keywords: Reaction kinetic, electrode, MEA

Introduction

PEMFC is a cell that it may produce the electrical energy by applying fuel. The principal operation of PEMFC is to convert the chemical energy by electrochemistry into the electrical energy. The components of PEMFC consists of bipolar plate of anode and cathode parts and the membrane electrode assembly (MEA) which possesses layers of anode, membrane and cathode. These five components are arranged into a stack known as so-called PEMFC [1,2]. From the point of chemical reaction, for one mol hydrogen under atmospheric pressure, temperature 287°K, the energy yielded as large as 237.200 J or equivalent to 1.23 V electric [1,3]. Practically, the PEMFC which applied non pure air, the humidity and hydrogen speed are well controlled, the maximum yielded cell voltan is found to be 1.16 V for open circuit. In stable condition it achieved 0.6 – 0.7 V and thus the PEMFC stack is large enough to yield 200-300 volt [3]. To reduce the MEA size, various techniques for instrumental manufacture have been employed for obtaining reduced thickness. For this purpose, Gulzow made a thin layer electrode by spraying method on the membrane surface [4]. By this method, the electrode thickness is found to be 10 until 5 micron [5] with a concentration of 0.05 to until 0.4 gcm^{-2}, the electrode thickness was obtained to be around 5 micron. According to Ralph, for a thin electrode, it can not assure a high MEA performance due to enhanced performance achieved only around 2% [6]. Hence, since 2002, investigators have started to improve the phenomenon of mass transfer in MEA. One of the popular manufacturing procedures was the patent of Wilson due to lower content catalyst [7]. The report of Chem stated that the enhancement of MEA performance achieved when the catalyst layer formed by thin layer method using ink [7,8]. The forming of catalyst layer was directly carried out onto the surface of GDL, but yielded not uniform thickness. In MEA investigation, the formation of catalyst layer thickness on the GDL surface gives effects to chemical reaction.

The underlying factors controlling fuel cell performance are best understood when one tries to break down a typical current/voltage curve into its contributions. As it turns out, these contributions can often be related to individual fuel cell components [8]. The scheme presented here is based on PEM fuel cell work. This seems fair because PEMFCs are currently the most widely studied type of fuel cell. Yet exactly the same primary performance parameters can be identified in all fuel cell types, so the scheme outlined here can be applied accordingly [9]. The smallest building block of a PEM fuel cell is the membrane electrode assembly or MEA, as shown in Fig. 1. Consisting of two electrodes, anode and cathode, and the polymer electrolyte, it essentially forms a complete fuel cell [10]. Before discussing current/voltage curves in greater detail, it is instructive to consider the respective functions of the MEA components. Figure 1 illustrates that the electrochemical reactions.

$$H_2 \rightarrow 2H^+ + 2e^- \ (E_r = 0 \text{ V}) \qquad (1)$$
$$1/2\, O_2 + 2H^+ + 2e^- \rightarrow 1H_2O \ (E_r = 1.23 \text{ V}) \qquad (2)$$

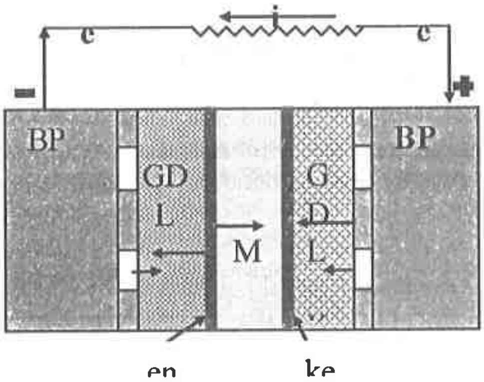

Figure 1. The element of MEA

Take place at the anode and the cathode catalyst layer, respectively. The gas diffusion layer (GDL) or electrode substrate (or electrode backing material) at the anode allows hydrogen to reach the reactive zone within the electrode [11,12]. Upon reacting, protons migrate through the ion conducting membrane, and electrons are conducted through the substrate layer and, ultimately, to the electric terminals of the fuel cell stack. The anode substrate therefore has to be gas porous as well as electronically conducting. Because not all of the chemical energy supplied to the MEA by the reactants is converted into electric power, heat will also be generated somewhere inside the MEA [13]. Hence, the gas porous substrate also acts as a heat conductor in order to remove heat from the reactive zones of the MEA.

At the cathode, the functions of the substrate become even more complex. Product water is formed at the cathode according to Eq. (2). Should this water exit from the electrode in liquid form (as it usually does if the reactants are saturated with water vapor), there is a risk of liquid blocking the pores within the substrate and, consequently, gas access to the reactive zone [14]. This poses a serious performance problem because for economic reasons the oxidant used in most practical applications is not pure oxygen but air. Therefore, 80% of the gas present within the cathode is inert with all associated boundary and stagnant [14].

layer problems. Fuel cell operation will therefore result in a depletion of oxygen towards the active cathode catalyst. The membrane acts as a proton conductor. As we will see, this requires the membrane to be well humidified because the proton conduction process relies on membrane water. As a consequence, an additional water flux from anode to cathode is present and is associated with the migration of protons (electro-osmotic drag). Since this will eventually lead to a depletion of water from the anode interface of the membrane, humidity is often provided with the anode gas by pre-humidifying the reactant.

Electrochemical kinetics

Figure 1 shows the volume element of MEA in PEMFC. In this element, there are three zones, i.e. the anode center, the polymer electrolyte membrane and the cathode center. The chemical reaction in the three zones is shown in the figure. To formulate the Influence of Electrode Properties on the Hydrogen Reaction Kinetic in Fuel Cell MEA, the hole diameter and GDL hole are assumed to be much higher than the hole diameter and electrode hole therefore the gas flow obstacle is larger than the electrode layer part. The left side of anode and the right side of cathode are assumed to be the collector of current density produced by chemical reaction [15]. The H2 humidity and the dry air are fixed for constant concentration. The water dropplets occured at anode and its transfer via cathode hole is neglected. The mixture of H2 gas and H2O in anode and O2 in cathode are followed the gas ideal rule [16]. The electrode is assumed as homogen medium possessed uniform holes. By this assumption, when operating on pure hydrogen, the anode stays at a potential close to the theoretical reversible potential of a hydrogen electrode, i.e., E_r=0 V-compare Eq. (1). In electrochemical terminology, this corresponds to a low over potential indicative of a kinetically facile reaction[10]. For the moment, we will therefore neglect the anode contribution towards the cell voltage, assuming an anode potential of 0 V. Since the cell voltage is the difference between anode and cathode potential, the cell voltage will be — to good approximation — identical to the cathode potential, *Ec*. In contrast with the anode reaction, the oxygen reduction reaction at the cathode is an activated process and therefore exhibits a much higher overpotential [17]. The Butler–Volmer equation, the key equation in electrochemical kinetics, gives a mathematical description of such activated processes [10,18,19,20].

$$i_c = i_0\left[\exp\left(\frac{\alpha_a F}{RT}\eta\right) - \exp\left(-\frac{\alpha_c F}{RT}\eta\right)\right]) \qquad (3)$$

The last exponential value in eq.(3) approached to zero [10], $\exp\left(-\frac{\alpha_c F}{RT}\eta\right) \cong 0$, therefore the polarization approximation electrochemical is as follows:

$$\eta = \frac{RT}{\alpha_a F}\ln\frac{i_c}{i_{o,c}^{ref}} \qquad (4)$$

The modeling of electrode activation

The electrode structure in MEA is determined by the thickness, hole, hole diameter, and active surface area [13,7]. The variation of the three parameters affects the current density. At a given thickness, hole, hole diameter and surface area, it will show a certain value [20]. The higher thickness value will show the lower hole. This case also occured for a given thickness that will show a certain hole diameter and active surface area [18]. To obtain the correlation between electrode properties and the hydrogen kinetic, the coefficient $\frac{RT}{\alpha_a F}$ in eq. (4) is modified into non dimensionless as shown below:

$$\eta = \frac{RTE_r}{\alpha_a F}\frac{1}{E_r}\ln\frac{i_c}{i_{o,c}^{ref}} \qquad (5)$$

Theoretically, the coefficient is influenced by the structure of electrode layer, i.e. the thickness, hole, hole diameter, and active surface area.

$$\frac{RTE_r}{\alpha F} = a_2\left(\frac{t_e}{W_1}\right)^{c_5}\left(\frac{dp}{W1}\right)^{c_6}(\varepsilon)^{c_3}(S_a W_1)^{c_7} \qquad (6)$$

An intercorrelation between thickness and hole diameter, hole, and active specific surface area [17,18,20] simplified eq.(6) and defined as follows:

$$\frac{RTE_r}{\alpha F} = a_1\left(\frac{t_e}{W_1}\right)^{c_1}\left(a_1\left(\frac{1}{a_o}\frac{t_e}{W_1}\right)^{1/x1^{x2}}\right)^{c_2}\left(a_2\left(\frac{1}{a_o}\frac{t_e}{W_1}\right)^{1/x1^{x3}}\right)^{c_3}\left(a_3\left(\frac{1}{a_o}\frac{t_e}{W_1}\right)^{1/x1^{x4}}\right)^{c_4} \qquad (7)$$

or

$$\frac{RTE_r}{\alpha F} = a_4\left(\frac{t_e}{W_1}\right)^{c_{10}} \qquad (8)$$

Furthermore the constant is introduced in eq.(5) therfore the influence of electrode properties on the hydrogen reaction kinetic which stated in the equation of kinetic performance is as follows:

$$\eta = a_4\left(\frac{t_e}{W_1}\right)^{c_{10}}\frac{1}{E_r}\ln\frac{i_c}{i_{o,c}^{ref}} \qquad (9)$$

To yield the various size of thickness, the electrode on the GDL surface using spraying. The movement with x-y coordinate is determined by the frequency (ω), nozzle height (W1), distribution distance (W2), the number sprayer at substrate (N) and the speed of nozzle movement (S). If the nozzle height W1 is direct proportional to Δx and W_1 is also required to be linear to W_2. Therefore, the nozzle movement is described as dimensionless number and defined in eq.(10).

$$N_{ukm} = \frac{(S - 2\omega y)}{2\omega W2} \qquad (10)$$

The CAD is adjusted with the dimension of *x, y, n, S* dan W_1. Moreover the nozzle position during manufacturing is controlled by adjusting with the parameter of N_{ukm}. Each value of N_{ukm} will yield electrode thickness on the GDL surface. The correlation of thickness is defined in eq.(11).

$$\frac{t_e}{W_1} = a_0 N_{ukm}^{\ x1} \qquad (11)$$

The size of thickness (t_e) is influenced by the spraying characteristic number (N_{ukm}). The combining of eq.11 and eq.9 of kinetic performance can be expressed in eq.12.

$$b = \frac{a_4(a_0 N_{ukm}^{\ x1})^{c_{10}}}{E_r} \qquad (12)$$

$$\eta = b\ln\frac{i_c}{i_o} \qquad (13)$$

Assuming an overpotensial in MEA □ $\eta = E - E_r$. This leads to the expression for the cell voltage, E_c: $E_c = E_r - b\ln\frac{i_c}{i_o}$

(14)

where E_r = reversible potential for the cell, b = Tafel slope for oxygen reduction. The so-called Tafel slope b is determined by the nature of the electrochemical process. For the oxygen reduction reaction [$n = 2$, in practical fuel cells, b is usually between 40 and 80 mV [10]. The main factor controlling the activation over potential and hence the cell potential, Ecell = E_c, is the (apparent) exchange current density i_0. Eq. 14 demonstrates that, due to the ln, a tenfold increase in i_0 leads to an increase in cell potential at the given current by one unit of b. It is important to emphasize this point [10.11]. While the Tafel slope b is dictated by the chemical reaction (and the temperature), the value for i_0 depends on reaction kinetics [21]. Therefore, the hypothesis of the influence of electrode properties on the hydrogen reaction kinetic in fuel cell is much affected by the electrode thickness or N_{ukm} from the spraying method. To demonstrate the influence on the performance of MEA, a serial of electrode manufacture was carried out on the GDL surface.

The MEA manufacture

To manufacture an electrode, Pt/C (20 %) powder, Nafion solution (5 %), and isoprophyl alcohol were applied. The ratio of Pt/C and nafion solution is made to be 0.7:0.3. The ratio of Platinum and water is made to be 1:20. These substances are mixed in a mixing tank. The mixing operation is carried out with a speed of 1200 rotations per minute for 10 minutes. The yielded ink is known as the so-called catalyst ink. After the ink spraying on the surface of GDL with a given N_{ukm}, the membrane nafion 117 is cleaned with 3 % H2O2 at 80°C for one hour to oxidize the organic matter. Moreover, it is cleaned again with boiled water for one hour. Then, cleaned again with H_2SO_4 5 wt % at 80°C for one hour. This procedure is carried out to remove the non organic material which possibly in the membrane. Furthermore, the membrane is washed several times with water at 80°C until perfectly cleaned. The layers are combined based on the configuration of gas diffusion anode – membrane – gas diffusion cathode (GDEA-M-GDEK) or to be assembled into unity. The hot pressing is carried out at a pressure of 50 kgcm^{-2}, temperature 130°C for 3 minutes. The product of this assembly is called as MEA. Moreover, the bipolar plate is combined with MEA the so-called PEMFC. The test of performance is undertaken with fuel cell test (FCT) Arbin instrument, USA. This station is operated based on the parameter value which its value similar used for optimization of mathematic modeling. The machine controlling is undertaken via computer using MITS_Pro software. The determination of the desired product and also the duration of cell operation are also being carried out before the cell test undertaken. During the examination, the data value are recorded and collected automatically by the MITS Pro software. Furthermore, the graphs may be created for analysis purpose.

Results and Discussion

Fig.2 shows the performance of MEA starting point which is referred to commercial MEA standard. The area of this MEA performance is possibly being undertaken in the investigation. The line shows the area border of the electrode investigation in this reserach. During the electrode manufacture, the spraying characteristic is expressed as given below:

$$t = 51.645\left[\exp\left(-0.55N_{ukm}\right)\right] \qquad (15)$$

The operation of the test instrument using the following data as given below:

Speed H_2 , cm^{3}pm = 0.0003
Concentration of H_2 for surface hole (C_{H2} = Y_{H2} x P_o / H_{H2}), mol/m^3 = 0.4200
Mol fraction of H_2 for surface hole = 0.0642
Mol fraction of H_2 for inner part of anode = 0.6000
Speed O_2 for surface hole, sm^{3}pm = 0.0006
Concentration of O_2 (C_{O2} = Y_{O2} x P_o / H_{O2}), mol/m^3 = 5.6000
Temperature back input H_2,°K = 35300
Temperature back input O_2,°K = 35300
Pressure of O_2, P = 2.5 10^5
Pressure of H_2, Pa = 2.5 10^5
Operation time, minute, = 30.00
Data input time, second, = 10.000
Concentration of H_2 in active layer (C_{H2}^{ref} = Y_{O2in} x P_o / H_{O2}), mol/m^2 = 0.4000

Concentration of O_2 in active layer (C_{O2}^{ref} =Y_{O2in} x P_o/H_{O2}), mol/m^2 = 0.6000
Change of current density in anode ($i_{o,a}$) , A/m^2 = 10000
Change of current density ($i_{o,c}$), A/m^2 (Hoogers,20) = 10.000
Concentration of O_2 in active layer (C_{H2}^{ref} = Y_{O2in} x P_o / H_{O2}), mol/m^2 = 0.4000

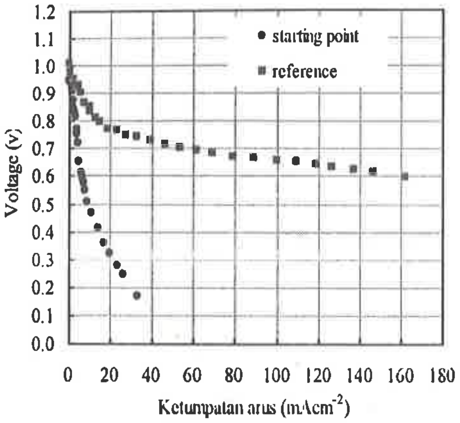

Fig. 2 Area of electrode investigation

At previous investigation, the electrode thickness very much affected the MEA performance [16]. The thinner electrode made the MEA performance elevated. This case due to gas reaction to produce the best proton and electron near the membrane surface [17,19]. However, at certain thickness, the performance will reduce because the reaction required enough electron [22]. To demonstrate this case, a configuration was made by making electrode on the anode surface as described in eq.(16).

GDEA$_x$(N_{ukm}) (16)

The x in the configuration of making A describes the number of electrode research in the anode part and the N_{ukm} is the number that shows the various spraying movement that produced anode thickness on the surface of GDL. In this investigation, the N_{ukm} value is plotted starting from 0.5 to 1.75. The stating point undertaken in this investigation applied a carbon cloth which is well prepared for hole as well as surface using Cabot and PTFE. This procedure caused the starting point of this investigation GDA 1(0.5) different from the starting point Fig.2. The results are shown in Fig.3. In Fig.3, the thinner electrode or the higher N_{ukm} value from 0.5 to 1.75, will yield the higher MEA performance. For N_{ukm} 1.75 will yield the highest MEA performance. Eq.15 is substitued into eq.14 to obtain the cell voltage, E_c:

$$E_c = E_r - \frac{a_4(25.822\exp(-0.55N_{ukm}))^{c_{10}}}{E_r}\ln\frac{i_c}{i_o} \quad \text{... (17)}$$

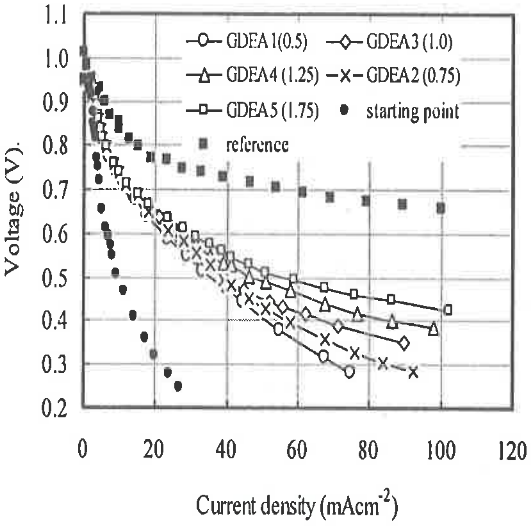

Fig.3 The performance of elctrode layer Ifb is simplified, so E_c :

$$E_c = E_r - c_{11}(\exp(-0.55N_{ukm}))^{c_{10}}\ln\frac{i_c}{i_o} \quad \text{... (18)}$$

$$b = \frac{RT}{\alpha F} = c_{11}(\exp(-0.55N_{ukm}))^{c_{10}} \quad \text{... (19)}$$

Eq.(19) is a Tafel slope that the value is determined by c_{10} and c_{11}. Value c_{11} shows the effect of condition values that not put in the theory of B-V arrangement. This two values can be determined by Hooke Jeeves calculation using basic program. By this calculation, the Tafel slope value of each examination can be determined as listed in Table-2

Table 1 lists that the thinner electrode or the higher N_{ukm} value, therefore the Tafel slope value getting lower. Graphically, the change of Tafel slope from the whole experiment is shown in Fig.4.

In this investigation, the reduced thickness caused the reduced Pt-loading. The reduction is found to be from 0.4 to 0.28 gcm^{-2}. The previous approach, if the mass-transport losses do not increase as the Pt-loading is reduced, the change in cell voltage as a function of anod Pt-loading can be describe mathematically on the basis kinetics [8,10]:

$$-b = \frac{RT}{\alpha F} = \left.\frac{\partial E_c}{\partial \ln[t_c]}\right|_{T,i} \tag{20}$$

Table 2 Tafel slope values by calculation.

Configuration of electrode preparation GDEA$_x$(N_{ukm})	Spraying movement N_{ukm}	thickness t_e (micron)	Tafel slope $b = \frac{RT}{\alpha F} = c_{11}(\exp(-0.55N_{ukm}))^{c_{10}}$ (Vdecade^{-2})
GDEA$_1$(0.50)	0.50	39	0.565
GDEA$_2$(0.75)	0.75	34	0.522
GDEA$_3$(1.00)	1.00	29.	0.511
GDEA$_4$(1.25)	1.25	25	0.471
GDEA$_5$(1.75)	1.75	20	0.435

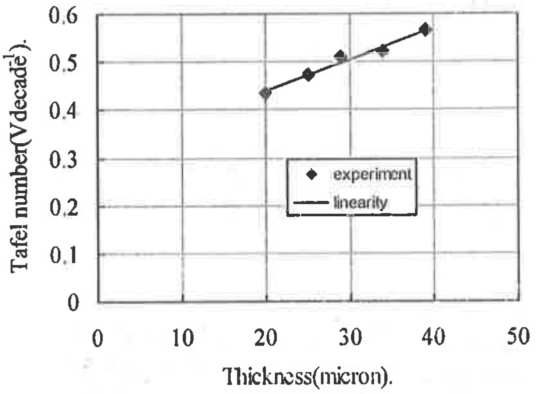

Fig. 4. The change of Tafel Slope

where t_c denotes the catode Pt-loading. Eq.(20) states that the change in cell voltage with the logarithm of the cathode Pt-loading (assuming the same catalyst is used) is proportional to the Tafel-slope. Eq. (20), again, assumes that H2/air reactants is not changed by reducing the Pt-loading pada, which was shown to be the case for a two-fold loading reduction (from 0.4 to 0.2 mgPtcm^{-2}). For a Tafel-slope of 70 mVdecade^{-1} at 80C, a loading reduction is thus predicted to lead to a voltage loss across the entire current density range of 20 or 40 mV, respectively. In this investigation, the examination was about the change in Pt-loading for anode. At the change in thickness from 39 micron to 20 micron or from 0.4 to 2.8 gcm^{-2}, it will show 0.56 and 43 V.

$$-b = \frac{RT}{\alpha F} = \left.\frac{\partial E_c}{\partial \ln[t_c]}\right|_{T,i} \quad \ldots (21)$$

$$-b = \frac{RT}{\alpha F} = \left.\frac{\partial E_c}{\partial \ln[51.645[\exp(-0.55N_{ukm})]]}\right|_{T,i} \quad \ldots (22)$$

This event shows that the change of thickness at cathode is more influenced than the change of thickness at anode. In this investigation, the experiment was conducted under room temperature that resulted lower Tafel slope value.

Conclusion

The hypothesis of electrode properties on the hydrogen reaction kinetic in fuel cell is much influenced by the electrode thickness or N_{ukm} by spraying method. From the discussion of the thickness influence, the tinner electrode or the higher N_{ukm} will yield lower Tafel slope value and apparently there is no any enhancement. At H2/air reactants is not changed by reducing the Pt-loading, which was shown to be the case for a two-fold loading reduction (from 0.4 to 0.28 mgPtcm^{-2}). For a change from 39 mikron to 20 micro or from 0.4 g/cm2 to 2.8 gcm^{-2}, it will show values of Tafel slope of 0.56 and 43 V.

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