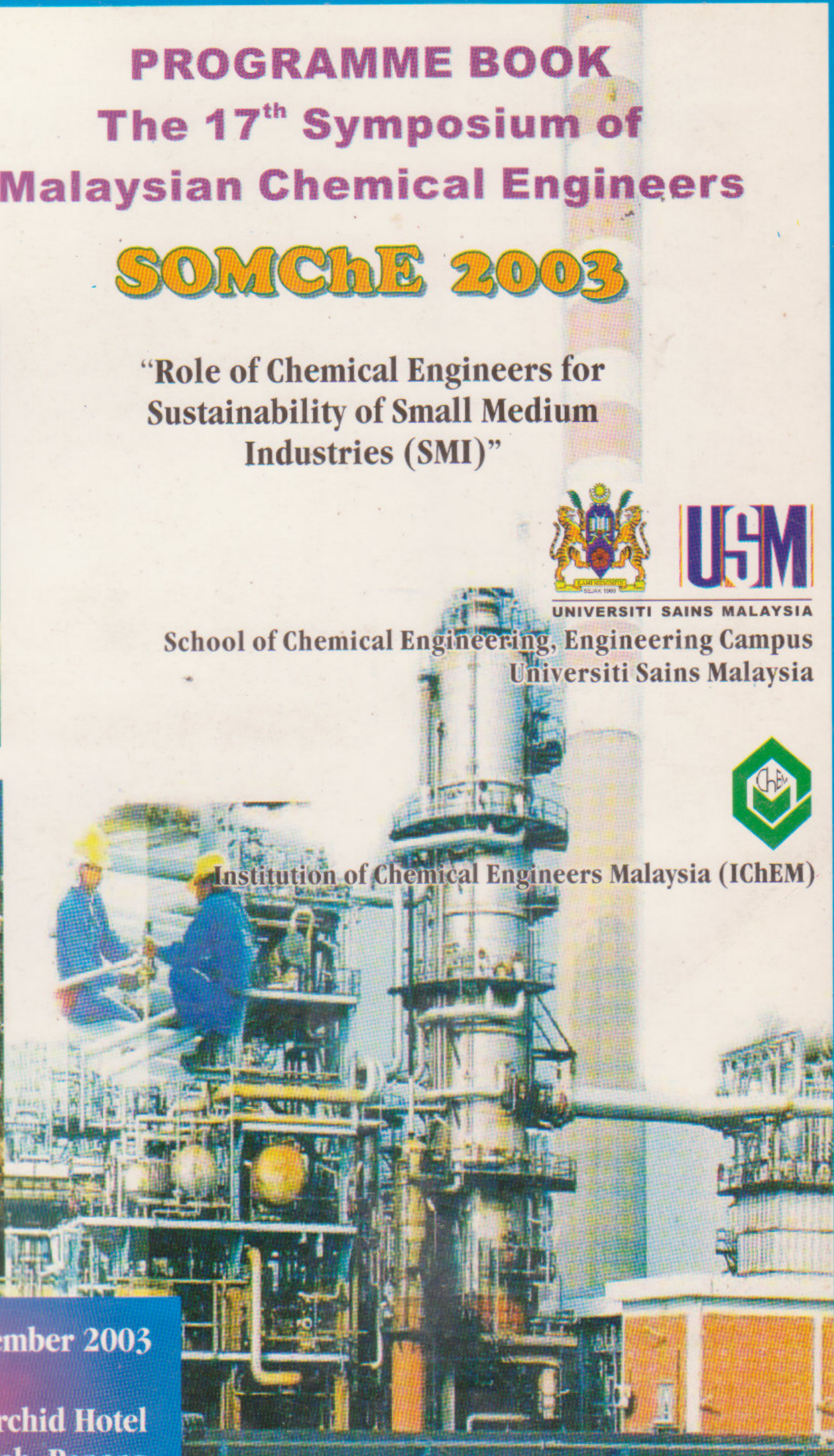


PROGRAMME BOOK
The 17th Symposium of
Malaysian Chemical Engineers

SOMChE 2003

**“Role of Chemical Engineers for
Sustainability of Small Medium
Industries (SMI)”**



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School of Chemical Engineering, Engineering Campus
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Institution of Chemical Engineers Malaysia (IChem)

29 - 30th December 2003

**Copthorne Orchid Hotel
Tanjung Bungah, Penang.**

**The 17th Symposium of Malaysian Chemical Engineers
(SOMChE 2003)**

**PROCEEDINGS:
SOMChE 2003**

THEME:

Roles of Chemical Engineers for Sustainability of Small
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Jointly Organised:



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Institution of Chemical
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PREFACE

SOMChE 2003 is the seventeenth in the annual series of Symposium of Malaysian Chemical Engineers. School of Chemical Engineering, Universiti Sains Malaysia and Institution of Chemical Engineers Malaysia jointly organize it with the theme "Role of Chemical Engineers for Sustainability of Small Medium Industries (SMI)".

Malaysia has transformed from a commodity-based producing nation to being a manufacturer of industrial products, geared towards exports. The small and medium industries (SMIs) play a critical role in the country's industrialization program. Therefore, such emphasis demands more development in research and development (R&D) and technological advancement. The sustainability of a SMI's competitiveness is very much dependent on the ability to continuously improve the productivity and quality of products and services. Productivity is never an accident, but it is always the result of a commitment to excellence, intelligent planning and focused effort. We as Malaysian chemical engineers have come together with a purpose to develop continuous effort to get SMI's to improve themselves, focus on developing a learning culture either by initiatives, coercion or jointly to get the SMI's to renew their skills so that they stay renewable.

The program features two keynote and 165 regular symposium papers. The papers are categorized in the following topics:

1. Process Control, Modeling and Simulation
2. Reaction Engineering and Separation process
3. Fundamentals
4. Bioprocess/Biochemical Engineering
5. Environmental Engineering
6. Alternative Energy
7. General

On behalf of the Organizing Committee, I would like to express our deepest appreciation to all authors and co-authors in taking time to take part in this conference for the betterment of SMI's. Any unforeseen errors during editorial process are greatly regretted. However, the information and opinions presented in the proceedings are entirely the responsibility of the respective authors.

Special thanks to all staff and postgraduates from School of Chemical Engineering, Universiti Sains Malaysia for their endless support, help and assistance in compiling and preparing this proceeding.

We believe the information shared in this proceeding will enlighten and accelerate the development of Small Medium Industries in Malaysia.

Dr Norashid Aziz
Secretary [SOMChE 2003]

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Diffusion Coefficient of the Dissolved Hydrogen Gas inside the Mesoporous Gas Diffusion Layer of MEA

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ABSTRACT

A mesoporous model was developed for the gas diffusion layer (GDL) electrode in order to increase the distribution of hydrogen gas into electrode layer of Membrane Electrode Assembly (MEA) of the PEM Fuel Cell. This model follows the concept of Fick's law to determine the flow of gas within the system. The flow gas distribution parameter is the diffusion of dissolved gas under the influence of temperature, radius of pore and the molecular weight of gas concerned. From the result obtained for the respective diffusion coefficient, the voltage and the current flow in the electrode can be determined through simulation using PEM Lab software. The best simulated voltage and current density results will then be used as an indicator in the fabrication of the GDL electrode. The fabrication of the electrode used in verifying the results of this model was performed using the casting technique. The measurement of gas distribution coefficient was done using BET instrument. The simulation result obtained showed that the gas diffusion coefficient distribution inside the GDL follows Fick's law with the dissolved hydrogen gas of $4.3 \times 10^{-6} \text{ m}^2\text{s}^{-1}$. Results of the experiment via the GDL mesoporous model showed the pore diameter of 31.78 \AA , pore volume of 0.09 cc g^{-1} and surface area of $253 \text{ m}^2 \text{ g}^{-1}$. The voltage and current density of the observed GDL are 1.05 V and 1 Amp/cm^2 , respectively. The performance of MEA using observed experimental GDL was compared with GDE from E-TEK and other commercial MEA to be similar.

Keywords: Mesoporous; Diffusion coefficient

1.0 INTRODUCTION

The MEA used in the fuel cell consists of various layers, such as, GDL, GDE, membrane, with different values of parameters. These values prior to fabrication need to be determined and optimized using simulation method. Since diffusion coefficient is the most sensitive parameter in fabricating the MEA, special attention has been focused to look into this issue from this aspect. Gas diffusion coefficient is dependent on the porosity of the GDL. With higher porosity, the performance of MEA will be increased due to the increase of mass transfer at the structure MEA (Nordlund, 2001, Hsin-Sen Chu, 2003). Sei (1999) reported that macroporous model has an ideal homogeneous mass transfer and is appropriate to be used in MEA (Sui, 1999, Sukkee, 2000), and the microporous type also produce high current (Lean, 2002). Based on the above studies, we have tried to observe the increase of performance of MEA by observing the mass transfer concept at GDL porous. Ruthven (1997) reported that the microporous, mesoporous and macroporous models of carbon particles have diffusion coefficient that depends on pore diameter of porous. Since microporous and macroporous models of GDL have been observed earlier by Lean and Sui, the objective of the present study is to determine the gas diffusion coefficient inside the GDL with mesoporous model. Then the value of gas diffusion coefficient will be used to predict the performance of MEA with mesoporous GDL model and compared to that of the microporous and macroporous GDL.

2.0 METHODOLOGY

In this model, we assumed GDL as a thin slab. Gas will diffuse under the influence of a gradient of pressure inside GDL and no convection of gas flow. The mass transfer of gas through GDL will follow the unsteady state process. The dynamics of gas transport through GDL barrier will be based on Fick's law as shown in equation 1.

$$-D \frac{\partial^2 P(x)}{\partial^2 x} = \frac{\partial P}{\partial t} \quad 1$$

Eq. 1 represents a distribution of gas inside GDL where D is the diffusion coefficient, P is the pressure, x is the thickness of the GDL and t is the time. In order to apply the Knudsen diffusion flow, the pore radii is assumed to be very small, thus making the diffusion coefficient, D in the Eq. 1 to be

$$D = \frac{4}{3} d_p \frac{\varepsilon_p}{\mu} \left(\frac{RT}{2\pi M} \right)^{1/2} \quad 2$$

where d_p is the pore diameter, ε_p is the porosity, μ is the viscosity, R is the gas constant, T is the temperature and M is the molecular weight. By substituting Eq. 2 into Eq. 1 will give us the representation of gas distribution inside GDL as follows,

$$\frac{4}{3} d_p \frac{\varepsilon_p}{\mu} \left(\frac{RT}{2\pi M} \right)^{1/2} \frac{\partial^2 P(x)}{\partial^2 x} = \frac{\partial P}{\partial t} \quad 3$$

The boundary condition of Eq. 3 are set to be $t = 0, x = -L, P = P_L, x_1 = L, P = P_L$. At $t = \infty, x = -L, P = P_1, x = L, P = P_1$. In this case, the temperature, T will be constant. The diffusion coefficient of dissolved gas inside GDL is substituted using the result of Hooke-Jeeves. The result will be compared with gas diffusion coefficient of Ruthven. In order to compare the simulation result we substitute the result of MEA for one stack equation and ran it using FEMLAB software.

3.0 EXPERIMENT

The materials used in fabricating GDL consist of Polimertetrafluoroethylene (PTFE) (Aldrich chemical Co, Inc) with 50% dispersed in the water, activated carbon (Ajax Chemicals), carbon cloth (E-TEK), 2-Propanol 99.5 % (Aldrich chemical Co, Inc), nafion solution (Dupont Chemicals) dan water. Activated carbon was used because it has high porosity to provide high electrical conductivity (Ralph, 2002), and also has high gas energy adsorption (Do and Wang, 1998). The mixing materials, called the carbon ink will be cast upon the carbon cloth using vacuum dryer with operating temperature set at 130°C for 4 hours. With this process a GDL composed of 28% PTFE and 72% activated carbon is formed. The porosity properties of GDL are measured using BET instrument via samples with size of 0.2 cm x 5 cm.

4.0 RESULTS DAN DISCUSSIONS

Table 1.0 shows that GDL has the following characteristics: pore diameter = 31.87 Å, volume of pore = 0.09 cc g⁻¹, and surface area = 253 m²g⁻¹. Based on Greff's classifications (1982), the pore size inside GDL falls in the region of mesopore. Meanwhile, the E-TEK GDL and commercial MEA (MEA COM) fall in the region of macropore. From DR method, the experimental prepared GDL has wide surface area compare to E-TEK GDE and MEA COM. Based on Ruthven, the GDL also falls under the mesoporous model. Therefore it can be concluded that observed properties of GDL is suitable to be used to develop GDE on the anode side of the MEA.

Table 1.0
BET Analyses of pore diameters and surface areas of MEA

Properties	GDL(EXP)	GDE (E-TEK)	MEA (COM)
Pore Diameter	31.87 Å ^o	68.36 Å ^o	86.70 Å ^o
Surface Area	253 m ² g ⁻¹	12.5. 7 m ² g ⁻¹	15.71 m ² g ⁻¹

4.1 Diffusion Coefficient of Dissolved Gas

The result from BET studies shows that the diffusion of hydrogen on GDL is to be carried out at 353 K and the pressure varies from 325 to 350 mmHg atm as shown in Figure 1. Using the parameters above, we could get a diffusion coefficient (D) of hydrogen dissolved gas inside GDL (normally called GDL(sim)) to be $1.43 \times 10^{-5} \text{ m}^2/\text{s}$ or GDL(sim) pore diameter as 33 Å. The experimental GDL (GDL(exp)) pore diameter obtained was 31.87 Å. The difference between GDL(sim) and GDL(exp) is relatively small for pore diameter and pressure as depicted in the figure. This means that the distribution of hydrogen dissolved gas inside GDL(exp) followed Eq. 3. On the other hand, the trend of the distribution of hydrogen dissolved gas inside GDL(exp) approaches the trend of the GDE(E-TEK). These trends approach each other, following Eq. 3, and the distribution of pore inside the GDL(exp) tends to approach GDE(E-TEK) as indicated in Fig. 2.

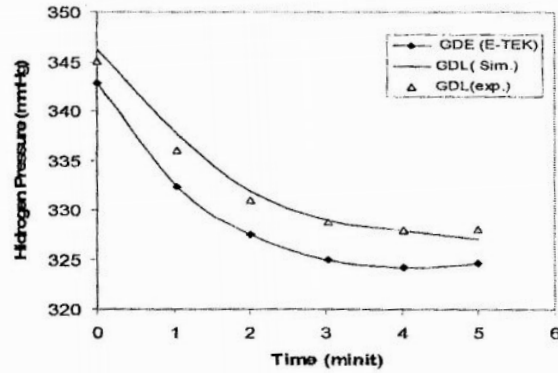


Fig.1. Dissolved hydrogen gas inside GDL(EXP)

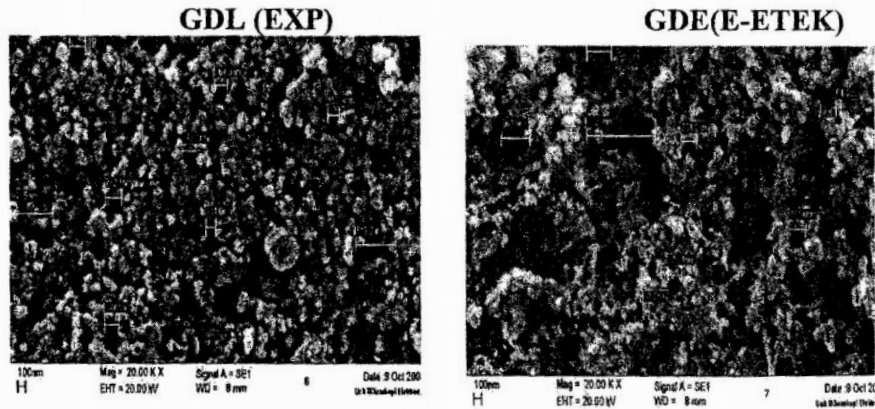


Fig.2. Comparison between the surface area of GDL(EXP) and GDE (E-TEK)

Comparison of the diffusion coefficients of hydrogen dissolved gas from these studies as well as from others showed that values obtained are different. The differences are due to the difference in the size of the pore diameter. The bigger the pore diameter, the higher will be the porosity of the hydrogen gas.

Table 2.0
Comparisons of diffusion coefficient for the GDL (EXP)

No.	Diffusion coefficient	Porosity Region	Source
1	$1 \cdot 10^{-6} \text{ m}^2/\text{s}$	Microporous	GDE (Lean, 2000)
2	$2.59 \cdot 10^{-6} \text{ m}^2/\text{s}$	Microporous	GDL(Sukkee, 2000)
3	$1.43 \cdot 10^{-5} \text{ m}^2/\text{s}$	Mesoporous	This study
4	$4.29 \cdot 10^{-5} \text{ m}^2/\text{s}$	Macroporous	This study
5	$5.49 \cdot 10^{-5} \text{ m}^2/\text{s}$	Macroporous	This study
6	$1.10^{-5} \text{ m}^2/\text{s}$	Macroporous	MEA (Sui, 1999)

4.2 Performance of GDL(exp) on MEA

In order to determine the performance of GDL(EXP), it is assumed that the porous model of GDL(exp) is similar to the porous model of GDE and the hydrogen flows continuously on MEA fuel cell and the fuel cell system has a perfect heat transfer to stabilize the temperature inside it. Using the assumption above, we could use the performance diagnostic equation of MEA fuel cell as suggested by Marr (1997). In the equation, the sum of current in a certain time and area of the GDL(exp) is a multiplication of diffusion coefficient of hydrogen dissolved gas and the changing of hydrogen pressure for a certain thickness of the GDL(exp) and the surface area. The value of current of MEA is strongly dependent on the diffusion coefficient of hydrogen gas. Using the operating conditions as shown in Table 3 for one stack fuel cell system and FEMLAB software, the voltage and current are determined and the performance of MEA could then be obtained as shown in Fig. 4.

Table 3.0
Data of GDL(EXP)

Characteristics of MEA	GDL (EXP.)	GDE (E-TEK)	MEA(COM.)
Operating pressure	1 bar	1 bar	1 bar
Operating temperature	80 °C	80 °C	80 °C
Dissolved hydrogen	5.19 mol/m^3	5.19 mol/m^3	5.19 mol/m^3
Dissolve oxygen	3.16 mol/m^3	5.19 mol/m^3	3.16 mol/m^3
Active layer thickness of the electrode	$480 \mu\text{m}$	$480 \mu\text{m}$	$466 \mu\text{m}$
Diffusion coefficient	$1.43 \times 10^{-5} \text{ m}^2\text{s}^{-1}$	$5.49 \times 10^{-5} \text{ m}^2\text{s}^{-1}$	$4.29 \times 10^{-5} \text{ m}^2\text{s}^{-1}$
Radius	$3.19 \times 10^{-9} \text{ m}$	$86.7 \times 10^{-9} \text{ m}$	$68.36 \times 10^{-9} \text{ m}$
Specific surface area	$2.8 \times 10^6 \text{ m}^2\text{m}^{-3}$	$3.12 \times 10^9 \text{ m}^2\text{m}^{-3}$	$3.14 \times 10^{10} \text{ m}^2\text{m}^{-3}$

In Fig. 3, the trend for polarization curve of the MEA performance using GDL(exp) has similar profile with the other GDE manufacturers. Lower value of hydrogen gas diffusion coefficient results in higher current of the MEA. Each porosity region shown in Table 2 will produce different value of current. It is shown that GDL(E-TEK) produces the current of 0.65 A/cm^2 , while MEA(COM) exhibits 0.91 A/cm^2 and GDL(exp) gives 1.0 A/cm^2 . We could conclude then from Fig. 3 that GDL(exp) will yield higher current for fuel cell. It means that the diffusion coefficient has very strong influence on the performance of MEA.

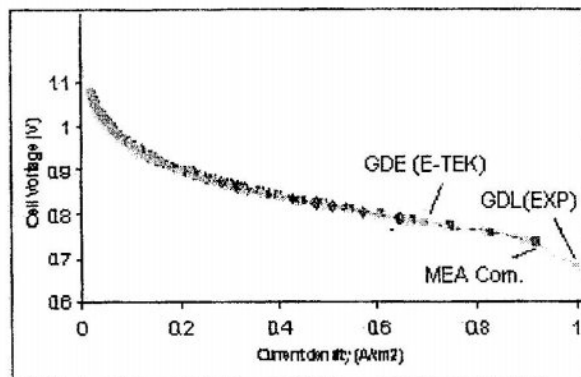


Fig.3. Performance model Characteristics of MEA Fuel Cell

6.0 CONCLUSION

We could conclude that the mass transfer of hydrogen gas inside the GDL(EXP) follows the Fick's law. The value of the diffusion coefficient of GDL has strong effects to the current produced in the fuel cell. With higher value of diffusion coefficient, the produced current in the fuel cell will also be increased. The voltage and current produced by GDL(exp) are 0.6 V and 1 A/m², respectively. Therefore, the GDL anode of the MEA could be prepared using the conditions as stipulated above.

7.0 ACKNOWLEDGEMENTS

The authors would like to express their gratitude to the UKM and MOSTE for the financial support through IRPA grant: IRPA 02-02-02-0003-PR0023/11-08

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