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DAFTAR ISI

Fabrication PEMFC based on the MEA design form

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Abstract

One of the working process in engineering of Proton Exchange Membrane Fuel cell (PEMFC) is to manufacture various MEA design. Among those designs were found the best one and being manufactured. This project introduces a spray robotic instrument with x-y axis configuration to manufacture the MEA design. The form of MEA layer which can be produced by this sprayer will follow the periodical function, while the layer included thickness, porosity, pore diameter, activated specific surface area untuk digunakan menyelenggarakan reaksi kimia dalam menghasilkan tenaga listrik. Layer sizes are expressed by sprayer character numbers. The number is a function of a robotic frequency value, traverse nozzle and x-y substrate dimension. The sprayer resulting two forms of MEA design will be close to the MEA commercial. Performance MEA design form yang diusulkan akan ditingkatkan malalui the control of layers fabricating using robotic characteristic numbers with typical nozzle configuration as a contribution to determine layer of MEA.

Keywords: Reaction, Current density, MEA design, fabrication, Number sprayer, Fuel cell

Introduction

One of the working process in engineering of MEA is to manufacture various design. Among those designs were found the best one and being manufactured. In manufacturing PEMFC there are eleven layers involved with the desain and manufacturing. The sequence of those layers is the bipolar plate (BP), gas flow field (FF), gasket (G), gas diffusion layer (GDL), electrode (E), membrane (M). This layer configuration is the BP-FF-G-GDL-E-M-E-GDL-G-FF-PP. The electrode assembly (MEA) part has 5 layers of GDL-E-M-E-GDL and recently was develop as 6 layers and called MEGA or G-GDL-E-M-E-GDL-G. Design and manufacturing of gas flow field PEMFC have fast growing development and manufacturing using CNC instrument. The MEA design begin to develop continuously to improve the performance of PEMFC. One of the basic principle to design MEA is to use nanotechnology so as to employ a very sensitive instrument such as vacuum plasma spray coating, vacuum atomization and sputtering.

Many simulations and experimental MEA previously undertaken to guide the explanation of layer size of the MEA in fuel cell and always to be developed by the more accurate equation [1]. Layer size of MEA and the interdependence between sizes is performed by graphics and the performance equation of PEM fuel cell [2,3,4]. From the mapping simulation and experimental results were carried out. these are the thickness, porosity, pore diameter activated specific surface area are the very crucial parameters of MEA [5,6,7,8], and are expressed by the equation related to voltage and PEM fuel cell current [9,10,11]. The thickness of the catalytic layer must be able to conduct protons and their reactions (3,12,13,15]. High active surface area catalyst is also essential to conduct an enough high reaction rate [8]. Besides that, the effective porosity will increase the current density of the PEM fuel cell flow [10]. Maximizing the current density is often carried out by adjusting the thickness and effective porosity to create uniform distribution of gas and humidity in MEA [3,4]. Generally, electrode sizes are optimized on the requirement of the tranfer of the mass, electrones, protons, and completing the ionic reaction on the electrode [5,6,7]. It seems that the best solution is to minimize the thickness of the catalyst layer while optimizing the combination of the electrode size parameters [9,10,11]. The layers preparation includes involves ink, coating, pressing and drying [1,2,5]. In general, the layers *preparation of MEA have to be carried out by utilizing the ink coating method on the* surface of gas diffusion electrode (GDE) or above the membrane surface [5]. One of the equipments required to perform the experiment is a spraying method. In this research the equipment used is Robotic Nozzle Sprayer which is able to conduct the uniform ink mixing with high tubulence. Besides that, it has a function as ink distributor to the surface of substrate. The objective of this research is to study the control of layers fabricating using robotic characteristic numbers with typical nozzle konfiguration as a contribution to do the design GDE and to determine electrode layer of MEA. Characterization. Benchmarking polarization of PEM fuel cell

Theory

The robot used in the system employs a specific attitude expression of the x-y configuration shown in Fig 1.

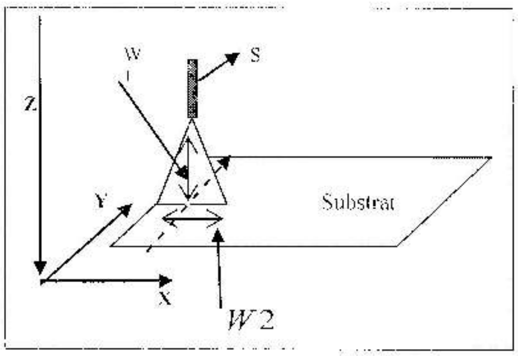


Fig 1. The x-y configuration

The spray variable is expressed by frequency (ω), nozzle height (W_1), distribution distance (W_2), division number of spray coating line on substrate (n) and nozzle velocity (S) [22]. The spray direction coating process is designed perpendicular to substrate. The nozzle frequency is given by equation 1.

$$\omega = \frac{nS}{2(ny + x)} \quad \ldots\ldots 1$$

In equation 1, the robotic frequency depends on S and N at certain boundary condition.. If the nozzle height W_1 is direct proportional to Δx and W_1 is designed to be proportional with W_2. Assumed the N_{spray} is known as the characteristic number of robotic spray, therefore the calculated value will be determined as follows based on the passing sprayer movement:

$$N_{spray} = \frac{S - 2\omega Sy}{2\omega W_1} \quad \ldots\ldots 2$$

Based on the equation 2, CAD is designed to possess the dimensions of x, y, n, S and W_1 level. The nozzle position on x-y axis is generated on the CAD system according to parameter n. The spray coating consists of the nozzle position and control code. The control code has a value corresponding to the desired substrate condition.

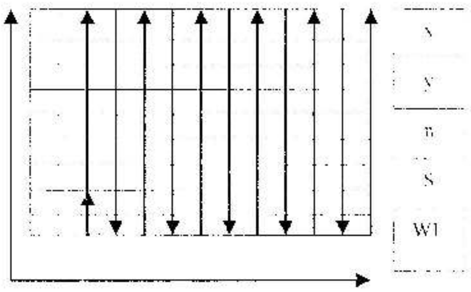


Fig 2. CAD of workpiece

Selain itu menghasilkan thick size (t_e), pore diameter (d_p), porosity (ε), typical activated specific surface (a_s). The variable of ink drop distribution of the nozzle will affect the layer size are μ , ν and p.

$$t_e, d_p, \varepsilon, a_s = f(K, \mu, \nu, \sigma, p) \quad \ldots\ldots 3$$

Assume p is constant, surface tension (σ) constant, thus theoretically the correlation of t_e, d_p, ε *and* a_s toward all variables and the robotic movement as well as the drop variable are given by dimensionless equation 4

$$t_e, d_p, \varepsilon, a_s = f(N_{spray}, Re, W) \quad \ldots\ldots 4$$

The viscosity effect and surface tension are constant and neglecting the solidifying effect on substrate surface. Based on equation 3, the t_e, d_p, ε, a_s are given by the dimensionless equation 4 to 5, as follows :

$$t_e, d_p, \varepsilon, a_s = f(N_{spray}) \quad \ldots\ldots 5$$

Equations 4 to 5, the t_e, d_p, ε and a_s of an electrode can be determined by the robotic characteristic number (N_{spray}). Assumed the layers results from robotic sprayer is set to be the MEA, therefore the relationship of N with the current density of PEM fuel cell can be formulated using Grujicic (2004) [18]. The spraying technique model for MEA fabrication is as follows:

Current density (i):

$$i = K_1 C_{O2} (1 - (K_5 \exp(-K_6(\phi_s))^{1/2}) \ldots$$
$$x \coth(K_5 \exp(-K_6(\varphi_s)))^{1/2} \quad \ldots\ldots 6$$

$$K_4 = \frac{12t_c(1-\varepsilon)FD_{O2}}{0.5d_p} = f(N_{spray})\ldots\ldots\ldots\ldots 7$$

$$K_5 = \frac{i_{0,c}a_{s1}(0.5d_p)^2}{4FC_{o2}^{ref}D_{O2}} = f(N_{spray})\ldots\ldots\ldots\ldots\ldots 8$$

and Voltage (V):

$$V = E - \frac{RT}{0.5F}\ln\{f_1(dp,\varepsilon)C_{O2}(1-(f_1(dp,\varepsilon)\ \ldots$$
$$\frac{i_{0,c}}{C_{o2}^{ref}}\exp(-\frac{RT}{0.5F}(\phi_s))^{1/2})x$$
$$\coth(f_i(dp,\varepsilon)\frac{i_{0,c}}{C_{o2}^{ref}}\exp(-\frac{RT}{0.5F}(\varphi_s)))^{1/2})/i_{o,c}\ \ldots\ldots\ldots 9$$

[17,18,19,20]

Based on equations, the value of coeffisient model of current density depend on N_{spray}. The N_{spray} becomes the main control for manufacturing layer size of MEA design form

Generally, the MEA design form configuration employed by researches as shown in Fig.3a is the G-GDL-E-M-E-GDL-G. In this paper the catalyst layer employed the configuration as shown in Fig.3b to obtain highly activated specific surface area. In Fig.3c the catalyst utilised a support to composite the catalyst into the membrane (novel).

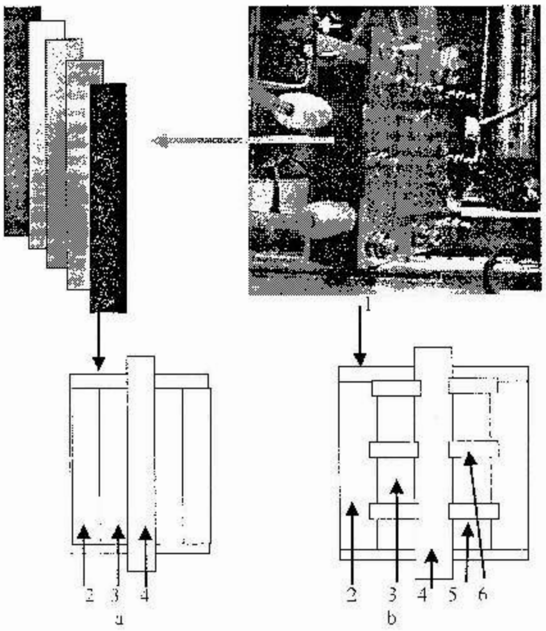


Fig.3. The MEA design configuration :(1) Gasket, (2) GDL, (3) electrode, (4) membrane, (5) composite, (6) electrode support

Experiment

Gas Diffusion Layer (GDL) in MEA employed Vulkan XC-72R carbon black with particle size 38 nanometer or aggregate 5 micron, PTFE 60 %(w%), prophyl alkohol 89 % and carbon cloth with a thickness of 210 micron. Vulcan XC-72 loading in GDL is as large as 5 mg/cm2 and PTFE 1.25 mg/cm2. The electrode layer in MEA employed a mixture of Pt/C(20 %), nafion solution (5%), prophyl alkohol 89 % and water. The weight ratio of Pt/C and Nafion in electrode ink was made to be 0.7 : 0.3 [20]. The ratio of Nafion with water was made to be 1:15. To obtain a sprayer characteristic, Pt/C ink was spread over the GDL surface with N values of 0.5 to 2.0. The spraying pressure of air mixture was 4 bar. The spraying applied hot plate under 60 °C to remove isoprophyl alcohol.

The flow chart of spraying is shown in Fig. 4 with operational condition as follows : nozzle height 7, hot plate under 60°C, air compressor 4 kg/cm^2 [21,22]. The pattern of nozzle 7 is shown in Fig. 2. The mixing process in mixing tank 5 is set to be laminair. The characteristic number of spraying is adjusted by PLC panel as desired. When the temperature of GDL achieved 60°C, then the speed of ink catalyst is set as desired. At the same time, the robotic is set to be on. After spraying process, the substrate should be dried for 1 h and then put in the dryer under 80°C for 3 h.

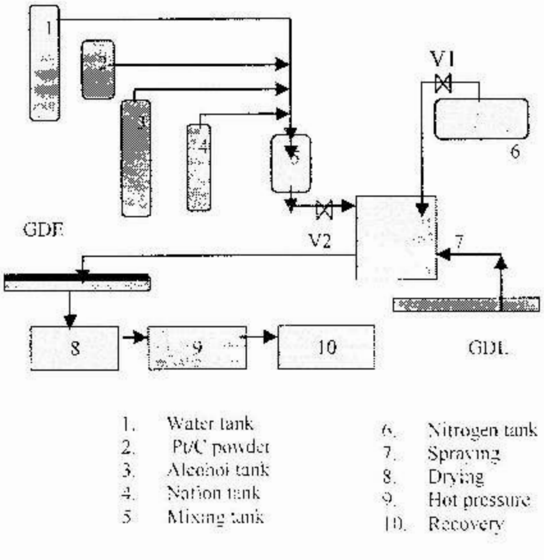


Fig. 4. The flow chart of GDL fabrication

Afterwards hot pressure should be undertaken for 3 minutes, under 130°C and pressure of 50 kgcm^{-2}. This kind of work has been done for various numbers of spraying characteristic. The next step, examinations have been carried out for thickness, holes, hole diameter, GDE surface area to obtain spraying data. The technique of GDE fabrication has been adjusted to the available design as well as the technique for characterization. The work has been done after selecting spraying variables in the characterization work. The same method has been done for GDE fabrication. The GDE fabrication are shown in Fig. 3a and 3b. After finishing GDE fabrication, the Nafion 117 membrane is cleaned by 3 wt % H_2O_2 under 80°C for 1h to oxidize organic impurities. Impurities are removed by boiled water for 1h. To remove the expected inorganic in the membrane, the membrane should be cleaned by H2SO4 5 wt% under 80 °C for 1h. The membrane is washed by water under 80°C several times until really clean. Moreover, the layers are combined by configuration type of GDEA-M-GDEK. Hot pressing has been done under 50 kg/cm2, at 130°C for 3 minutes. The result of this arrangement is called as MEA. The instruments used for analysis to collect spraying data are the porosimeter and FCTS. Examination using porosimetry is employed for collecting data of holes, hole diameter and hole surface area.

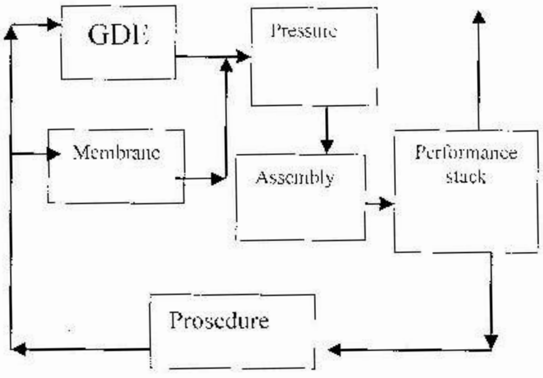


Fig. 5. Prosedure MEA

Furthermore, the layer is merged with the membrane as shown in the flow chart in Fig.4. The MEA from hot pressing is examined by FCTS made of Arbin. For analysis of electro potential optimization in the spraying performance model the experiment applied VB.

Result and discussion

The research in MEA layer designing includes two steps i.e. the sprayer characteristic and MEA designing and manufacturing. The effect of spray ding on electrode layer size is demonstrated in Fig 6. From Fig 6 it will determine the electrode thickness on the surface of GDE. When we observe the distribution of data point there are many choice to determine electrode thickness which followed the method as described in journal. In this investigation, the robotic sprayer is adjusted to yield electrode thickness from 0.01 mm to 0.05 mm [20]. The correlation of porosity, diameter pore and activated specific surface area with N is illustrated as exponential function. For porosity, it has a coefficient of 2.572 10^{-5} and power of -0.59. For hole diameter, it has coefficient of 3.87 10^{7} and power of 0.23, and for activated specific surface area it has coefficient of 3.84 10^{5} and power of 0.094.

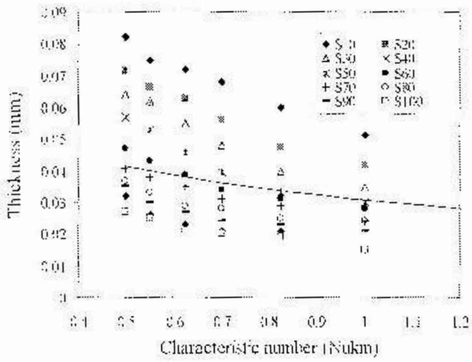


Fig 6. The effect of characteristic number on electrode thickness

We obtain the interdependent of sizes, i.e. t_e, d_p, ε, a_s which one another will be related with values of N_{spray}. The smaller the value of t_e while d_p, ε and a_s will be larger. This condition indicated that when one of the sizes to be determined so the other size will be fixed. As so far the characteristic sprayer will only adjust the thickness and forms a layer followed MEA design.

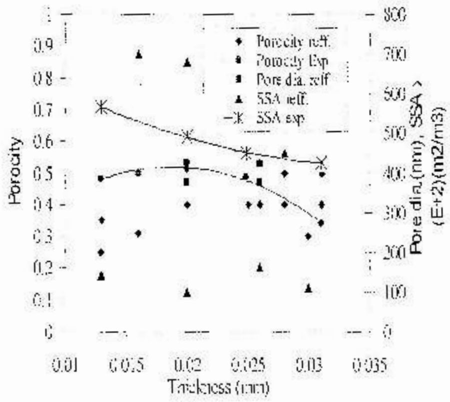
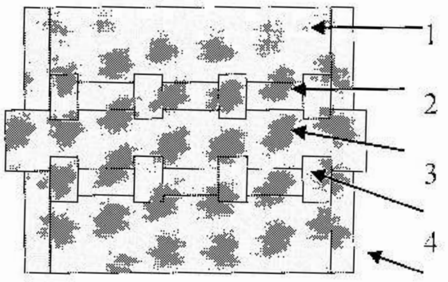


Fig.7. The mapping from sprayer result

The mapping from simulation modelling and experiment will show the thickness value of electrode (t_e) of the PEM fuel cell t_e that are from 10 to 35 micron (0.001-0.035 cm). ε from r 0.25 to 5. size average, d_p is 400 nm and a_s are from 1100 to 6000 cm^2cm^{-3} [19]. It is found that some values from eq. are located on that mapping. This implies that the sprayer can follow the results of simulation and experiment in journals. For example, the mapping from sprayer result are demonstrated in Fig.7. In Fig.7. the porosity point and activated specific surface area (SSA) obtained from sprayer are located in the mapping region, however the SSA still much below the "high performance specific surface area i.e. $1x10^7cm^2cm^{-3}$ [10]. The difference of mapping value with experimental result probably caused by the condition of drying treatment, hot pressing [5], and membrane treatment. On other side the robotic sprayer is not as accurate as CNC, vacuum plasma spray coating, spray vacuum atomization and sputering. However the sprayer already used in MEA design manufacturing. The following design uses electrode loading of 0.28 mgPt/cm2 and size as shown in Fig.8



1. GDL 210 micron
2. Catalyst = 20 micron
3. Membrane 180 micron
4. Electrode Support
5. Gasket 230 micron

Fig. 8. The MEA design form

In this design, the experimental MEA starting point, N_{spray} is to be 0.7. and will yield the current density of PEMFC around 60 mA/cm2. Afterwards the N_{rs} is to be 1.0, will produce current density of 85 mA/cm2. In turn it implies that thinner electrode on GDL surface, the current density obtained becomes larger. The thickness of the catalytic layer got thinner will be able to conduct protons and their reactions (3,12,13]. This is due to the higher active surface area catalyst obtained [8]. Besides that, the effective porosity will increase the current density of the PEMFC flow [10]. By adjusting the N_{rs} , the thickness and porosity will produce uniform distribution of gas and humidity in MEA layers [3,4]. It seems that the best solution to minimize the thickness of the catalyst layer as done in this research is in agreement with previous journals [9.10,11].

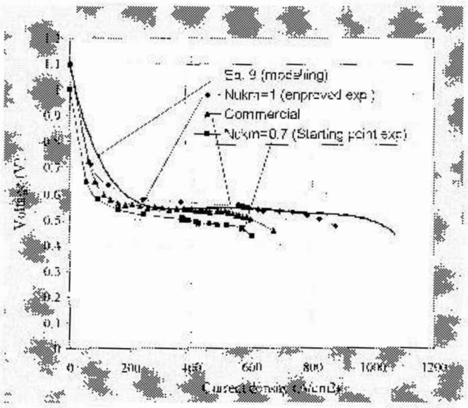


Fig. 9. The polarization of PEM fuel cell

In manufacturing the electrode layers there is an improvement in current density as large as 41.6% from the experimental MEA starting point mentioned above. The increment is high enough therefore the N_{spray} will be found to be 1 and used for manufacturing both for designing as is shown in Fig. 8 as well as other design.

Conclusion

The sprayer in this experiment can yield various sizes of thickness, diameter pore, porosity and activated specific suface area. Layer sizes can be performed with characteristic number robotic sprayer. Characteristic layer on substrate is close to characteristic simulation and experiment in journals.. The increment of the value of sprayer characteristic will yield the improvement of the PEM fuel cell performance.

From the experimental results, the value of N is to be unity and can improve the electrode performance. The configuration coating on substrate is a periodic function. The value of N is to be unity and can be utilized as the method for manufacturing MEA design.

Acknowledgement

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NOTASI

a_s = activated specific surface area. m2/m3
d_p = pore diameter. m
n = division number
N_{spray} = Number of robotic sprayer
p = pressure, bar
R_e = Renould number
S = speed nozzle. m/jam
t_c = thickness. m

v	= velocity ink , m/jam	
$W1$	nozzle height , m	
$W2$	= ink distribution width, m	
W_e	= Weber number	
X,Y	= substrate coordinate, m	
μ	= viscosity, cp	
ε	= porosity	
ω	= frequency, 1/second	

References

[1]. Ramli Sitanggang, Mohamad A.B., Daud, W.R.W., Kadhum, A.A.H., Iyuke S.E. Fabrication of gas diffusion layer based on x–y robotic spraying technique for proton exchange membrane fuel cell application. An Internatonal Journal, Energy Conversion and Management 50 (2009) 1419–1425

[2]. Iyuke, S.E., Mohamad, A.B., Khadhum, A.A.H., Daud, W.R.W., Rachid, C..Inproved membrane and electrode assemblies for proton exchange membrane fuel cells. Journal of Power Source 114(2003),195-202

[3]. Gurau,V., Liu, H., and Kakac, S.. Two-Dimensionnal Model for Proton Exchange Membrane Fuel Cells. Dept, of Mechanical Engineering, univercity of Miami, Coral Gables. (2002) Fl33124

[4]. Huang Z. Experiment and Matematical studies for PEM fuel Cell Performance, M.S. thesis, University Miami. coral gables. Florida., (2000)

[5]. Siegel, Siegel P.N. A two-dimensional computasional model of PEMFC with liquid water transport. Journal of Power sources, inpress (2003)

[6]. Paganin, V.A., Ticianelli, E.A., Gonzalez, E.R. Development and electrochemical studies of gas diffusion electrodes for polymer electrolyte fuel cell. J. Appl. Electrochem. (1996) 26297-304

[7]. Nam.J.H.,Kavianti M. Effective diffusivity and water-saturation distribution in single- and two-layer PEMFC diffusion medium". Pergamon. International journal of mass transfer 46(2003) 4595-4611

[8]. Hsin-Sen Chu, Chung Yeh, Falin Chen." Effects of porocity change of gas diffuser on performance of proton exchange membrane fuel cell. Journal of Power Sources 123(2003) 1-9

[9]. Tianhong Zhou and Hongtan Liu. Ageneral There-dimensional Model For proton Exchange Membrane Fuel Cell. J.Transport phenomena, 3(3):177-198(2001)

[10]. C.S.Kong, D.Y Kim, H.K. Lee. Y.G. Shul, T.H. Lee, Influence of -pore size distribution of diffusion layer on mas- transport problem of proton excange membrane fuel cell, J. Power Sources 108(2002) 185-191

[11]. Lixing You , Hongtan Liu, Aparametric study of the chatode catalyst layer of PEM fuel Cell using a pseudo-homogenous model. International journal of hydrogen Energy 26(2001)991-999

[12]. Andrew Rowe, Xianguo Li., Matematical modeling of Proton exchange membrane fuel cells., Journal of power sources 102(2001) 82-96

[13]. Shan-Hai Ge. Bao-Lian Yi.. A matematical model for PEMFC in different flow modes., Journal of Power Sources. impress (2003).

[14]. P.C. Sui and L.-D Chen, James P.Seaba. Yoshinori Wariishi. Modelling and Optimation of a PEMFC Catalyst Layer. International Conggress and Exposition Detroit. Michigan(1999).

[15]. Tien-Chien Jen, Tuanzhou Yan , Shih-Hung Chan.. Chemical reacting transsport phenomena in PEM Fuel Cell. International Journal of Heat and Mass Transfer 46(2003)4157-4168

[16]. Lixin You, Hongtan Liu. A two-phase flow and transport model for the cathode of PEM fuel Cell., International Journal of heat and mass transfer 45(2002)2277-2287

[17]. J.J. Baschck, Xianguo Li.. Modelling of polimer electrolyte membrane fuel cells with variable degrees of water flooding. Journal of Power Souece 86(2000) 181-106

[18]. E.Guzlow, T. Kaz, New Result of PEMFC electrode pruduce by DLR dry preparation technique , J..Powre sources 106 (2002)122-125

[19]. G. Maggio. V. Recupero, L. Pino. Modelling polymer electrolyte fuel Cell: an innovative approac., Journal of Power Source 101(2001) 275-286

[20]. M. Grujicic, K.M Chittajallu, Design and optimization of polymer electrolyte membrane (PEM) fuel cells. Applied Surface Sience inpress (2004)

[21]. Litster S., McLean G.. "PEM Fuel cell electrodes". Journal of Power Sources. inpress (2004)

[22]. E.A.Ticianelli, C.R. Derouin. A. Redondo. S.Srinivasan, Methods to advance technology of Proton exchange membrane fuel cell. J. Electrochem. Soc.135(1988)2209-2214

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