

Matching of critical parameters in a small non-pressurized non-humidified PEMFC stack

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Abstract

The desired performance of a proton exchange membrane fuel cell (PEMFC) stack is always hindered by improper matching of critical design and operating parameters associated with the stack. This paper presents typical design considerations for a small stack, which essentially operates under non-pressurized and non-humidified conditions. With the objective of maximized energy efficiency (or fuel economy), the relationships between conductivity and water loading, water loading and relative humidity, and relative humidity and air stoichiometric number were established under different temperatures. Through these relationships, the conductivity, which is performance related, is correlated with the air stoichiometric number. Thus, the latter has become the sole control parameter for the small PEMFC stack at a fixed temperature operating under the above mentioned constraints. The results show that, under a low air stoichiometric number, the higher the operating temperature is, the higher the conductivity will be. However, opposite trend can be seen for high air stoichiometric number. For a fixed operating temperature, the conductivity drops dramatically when the air stoichiometric number exceeds the threshold value. The higher the operating temperature, the more rapid drop of the conductivity against the increased air stoichiometric number will be.

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1. Introduction

Fuel cell systems produce electrical energy at very high efficiency. Generally, the efficiency is better than that of generators driven by internal combustion engines such as piston engines and gas turbines. Only large combined cycle power plants can outperform fuel cells. However, if high electrical efficiency is required, fuel cells can be combined with gas turbines, thereby surpassing the efficiency of combined cycle systems, while maintaining zero/low emission characteristics. In particular, due to the synergistic effects of the integrated solid oxide fuel cell and gas turbine technologies, simulation results showed that an overall system efficiency of 70% (net AC power to LHV of fuel, typically natural gas) or higher is possible with a more complex thermodynamic cycle [1]. In addition, due to the ability to integrate power production in dwelling

areas, efficient use of the waste heat for various applications is possible.

It is generally agreed that proton exchange membrane fuel cell (PEMFC) is the most promising fuel cell technology for mobile and portable applications due to its high energy density, low weight and volume compared with other types of fuel cells. In addition, its operating temperature of less than 80 °C makes it suitable for rapid start up. Literature review on the design of PEMFC showed that researchers either focus on detailed component and/or small stack design with the help of sophisticated models or by tedious experimental trial-and-error involving different conceptual design. Lee and Lalk [2] claimed to have developed a technique, which was specially designed so that models developed based on this technique can be used to determine the fundamental thermal–physical behavior of a fuel cell stack under any operating and design parameters. It can also be used to replicate the performance of a specific fuel cell system. Mann et al. [3] developed a generic model that could accept as input not only values of the operating variables such as anode and cathode feed gas, pressure and compositions, cell

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temperature and current density, but also cell parameters including active area and membrane thickness. A special feature of this model is the addition of a term to account for membrane aging by considering the deterioration of water-carrying capacity with time. Jiang and Chu [4] reviewed and evaluated the design of three types of PEMFC structure under different humidity and temperature, including bipolar, pseudo bipolar and monopolar stacks and concluded that they are suitable for moderate to high power, high power and low power (in particular with high voltage) applications, respectively. While sophisticated fuel cell models could give a detailed description of the fuel cell characteristics and performance subject to the change of performance related parameters or operating conditions, simple catalyst model developed by Chan and Tun [5] could provide the information on the relative effect of each key parameters on the fuel cell performance so that effort for improvement can be focused on. Rajalakshmi et al. [6] developed a technique for determining the distribution of current density in an operating PEMFC using segmented cell structure. Real-time current density distribution was measured which can contribute to an in-depth understanding of the reactant distribution over the active fuel cell area for optimization of the fuel cell performance. Kumar and Reddy [7] focused on the improvement of PEMFC performance through optimization of the channel dimensions and shape in the flow-field bipolar/end plates. The authors found that for hydrogen consumption greater than 80%, the optimum dimension values for channel width, land width and channel depth are close to 1.5, 0.5 and 1.5 mm, respectively. Their study also showed that triangular and hemispherical shaped cross-section channel performed better than square cross-section channel in terms of hydrogen consumption. Chan et al. [8] developed a PEMFC model with anode CO kinetics, which is essentially a model that marrying the work of Bernardi and Verbrugge [9] with that of Springer et al. [10]. The model estimates the superficial water velocity in the membrane electrode assembly (MEA) as well as the poisoning effect of CO concentration on the PEMFC performance. Hottinen et al. [11] studied a planar cell design for free-breathing fuel cell, in which the cathode side of the cell is directly exposed to the air, while the anode side flow-field geometry is of 'Z' shape. The authors claimed that this kind of design is feasible for small-scale applications, such as portable computers. Carnes and Djilali [12] presented an algorithm for estimation of PEMFC model parameters using a constrained nonlinear least square algorithm. Estimation of five model parameters (i.e., membrane conductivity, anode and cathode current density, oxygen diffusion coefficient in gas diffusion layer and catalyst layer) in a simple 1D electrochemistry model using two different experimental polarization curves has been demonstrated using this procedure.

Whatever approach the researchers adopted, the ultimate goal is to achieve improved PEMFC performance through optimization of key design parameters under the constraint of operational requirements. It seems that between tedious experimental trial-and-error and sophisticated modelling, there is a need to establish relationships for matching of critical parameters in the PEMFC stack. In this paper, the authors present typical design considerations for a small stack, which essentially

operates under non-pressurized and non-humidified conditions. With the ultimate goal of maximized energy efficiency (or fuel economy), the relationships between conductivity and water loading, water loading and relative humidity, and relative humidity and air stoichiometric number under different constant temperatures have been established. Through these relationships, the conductivity, which is performance related, is correlated with the air stoichiometric number. Thus, the latter has become the sole control parameter for the small PEMFC stack at a fixed temperature operating under the above mentioned constraints. Note that, since the focus is placed on small PEMFC stacks, the temperature range of 25–40 °C under study should be reasonable.

2. Non-pressurized, non-humidified, dead-ended anode flow channel PEMFC

A fuel cell that is an ambient pressure air-breather is designed to use low-pressure hydrogen on the anode side of the membrane electrode assembly and ambient air on the cathode side. This type of the cell can be designed to have a dead-ended anode flow channel with a pre-set purge cycle, which is sometimes referred to as burping the cell. A valve may be used to release a small amount of hydrogen from the anode flow-field in the other side of the flow channel where condensed liquid water and trace impurities of the fuel trapped in the anode flow channel could be scavenged. In this case, it can be assumed that all hydrogen introduced to the cell will be consumed and this consumption can be determined from the output current of the cell.

Water plays a critical role in proton exchange membrane fuel cells. Both the Nafion membrane and the Nafion material in the catalyst layers need to be hydrated in order to conduct protons efficiently. The protonic conductivity increases with the increase of water content. To achieve enough hydration, water is normally introduced into the cell externally by a variety of methods such as liquid injection, steam introduction and humidification of reactants by passing the hydrogen and air through humidifiers before entering the cell. Humidification by the last method is relatively easier to handle and therefore, it is the most commonly used technique. To simplify the system and to limit the amount of water to be carried with the fuel cell for the humidification purpose, the water generated by the fuel cell at the cathode should be recycled as much as possible.

The design of a fuel supply system for non-humidified and non-pressurised fuel cell is less costly. Since the cell is dead-ended, there is no need to meter or regulate the fuel flow rate. This means that no mass flow controllers are required, except one that is needed for measuring the fuel consumption. The cell consumes fuel as required from a source of low-pressure hydrogen. If there is no load connected on the cell, the fuel consumption is close to zero. This type of cell is self-heating up to the equilibrium temperature and is suitable for power output smaller than 100 W.

3. Model development

The protonic conductivity of a polymeric membrane is strongly dependent on membrane structure and membrane water

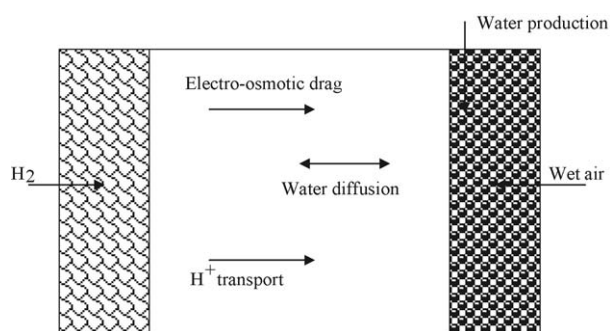


Fig. 1. Schematic of water sources and water fluxes.

loading or membrane hydration (λ). λ is defined as the number of water molecules per sulfonate acid groups.

$$\lambda = \frac{N(\text{H}_2\text{O})}{N(\text{SO}_3\text{H})}$$

Thus, a key challenge in the evaluation of ionomeric membranes for fuel cell has been focused on the analysis of combined structural and water uptake characteristics required to achieve the highest protonic conductivity.

Sources of water and the various flux components, which redistribute water in an operating PEMFC, are shown schematically in Fig. 1. Water is produced at the cathode by the electrochemical process and enters the cathode via the wet air as water carrier. Water is transported through the membrane from the anode to the cathode by electro-osmotic drag, i.e., by a water flux associated with the protonic current. The flux of water due to electro-osmotic drag ($\text{mol cm}^{-2} \text{s}^{-1}$) is given as:

$$N_{w,\text{drag}} = \frac{In_{\text{drag}}(\lambda)}{F}$$

where I is the cell current (A or C s^{-1}); $n_{\text{drag}}(\lambda)$, the electro-osmotic drag coefficient (cm^{-2}) at a given state of membrane hydration λ ; and F is the Faraday constant (C mol^{-1}). This water migratory flux tends to dry out the anode side of a cell under current and adds to the faradaic production of water at the cathode, thus producing excess water at the cathode. This build-up of water at the cathode is relieved by diffusion down the resulting water concentration gradient and by hydraulic permeation of water in differentially pressurised cells where the cathode is held at higher overall pressure. The fluxes ($\text{mol cm}^{-2} \text{s}^{-1}$) brought about by the latter two processes within the membrane are:

$$N_{w,\text{diff}} = -D(\lambda) \frac{\Delta c}{\Delta z}$$

$$N_{w,\text{hyd}} = k_{\text{hyd}}(\lambda) \frac{\Delta p}{\Delta z}$$

where D is the diffusion coefficient ($\text{cm}^2 \text{s}^{-1}$) in ionomer of water content λ ; $\Delta c/\Delta z$, the water concentration gradient (mol cm^{-4}) along the z -direction of membrane thickness; k_{hyd} , the hydraulic permeability (mol s g^{-1}) of the membrane; and $\Delta p/\Delta z$ is the pressure gradient ($\text{g cm}^{-2} \text{s}^{-2}$) along z . The steady-state water profile across the ionomeric membrane for given cell current density, external humidification conditions and differential pressurisation, is the resultant of electro-osmotic, diffusive and hydraulic fluxes.

Modelling work on the PEM fuel cell began as electrode–membrane–electrode models. Most previous models focused on the fully hydrated membrane fuel cell with the condition that reactants are thoroughly humidified prior to entering into the fuel cell. The membrane is liquid water-equilibrated membrane and the electrodes are hydrophobic in nature, which are made of the catalyst mixed with PTFE, then brushing liquid membrane solution onto the catalysed face of the electrode and allowing the liquid to seep into the electrode's pores. In this case, the conductivity is only a function of temperature. For example, the conductivity of Nafion is 0.17 S cm^{-1} at 80°C and 0.21 S cm^{-1} at 95°C [9]. However, for fuel cells that operate at low temperatures and reactants are not humidified or only slightly humidified, the membrane is water vapour-equilibrated membrane and the electrodes are usually hydrophilic ones, which are made of the catalyst mixed with liquid membrane solution. The behaviour of water vapour-equilibrated membrane is much more complex than liquid water-equilibrated membrane. In this case, the conductivity is a function of the operating temperature and airflow rate. The latter is applied to this study.

The conductivity of membrane is a function of water loading. If water loading is less than two, the Nafion membrane behaves like an insulator. Above this threshold, the conductivity rises dramatically with water loading, reaching a plateau in the semiconductor range of the order of about $10^{-1} \text{ S cm}^{-1}$ [13]. The water loading is, in turn, related to the activity of water (or the relative humidity), while the latter is, in turn, determined by the amount of water produced in reactions and the humidity of the air carrying to cathode. The amount of the air carrying to the cathode, in this study, is expressed as a function of air stoichiometric number and is defined as molar ratio of the actual inlet air supply to stoichiometric air in the H_2 –air reaction.

Since anode is dead-ended in this study, the air stoichiometric number and fuel cell operating temperature are the only two controllable parameters.

Fig. 2 shows the interdependency between any two key parameters that are related to the performance of PEMFC. Once each dependency is established, the relationship between the conductivity and the air stoichiometric number under a fixed operating temperature can be obtained.

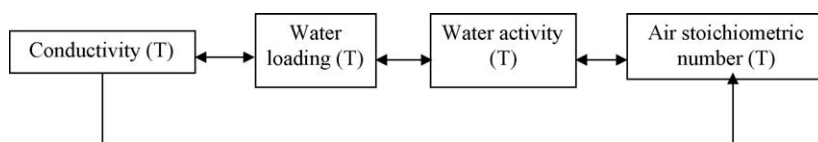


Fig. 2. Interdependency between key parameters under a fixed temperature.

4. Relationship between conductivity and water loading

For the case of a proton exchange membrane consisting of water as the solvent and hydronium ion as the protonated solvent, the conductivity model developed by the Thampan et al. [13] is expressed as:

$$\sigma = (\varepsilon - \varepsilon_0)^q \left(\frac{\lambda_1^0}{1 + \delta} \right) c_{\text{HA},0} \alpha$$

where ε is volume fraction of water in hydrated membrane; or wet porosity, ε_0 , the percolation threshold volume fraction of water in hydrated membrane, q , the Bruggemann or critical exponent; λ_1^0 , equivalent conductance for H_3O^+ at infinite dilution; δ , ratio of mutual to matrix effective diffusion coefficients; $c_{\text{HA},0}$, concentration of membrane acid groups; and α is the degree of acid-group dissociation. And

$$\varepsilon = \frac{\lambda}{\bar{V}_M/\bar{V}_2 + \lambda}$$

$$c_{\text{HA},0} = \frac{1}{\lambda \bar{V}_2}$$

where λ is water loading, or the number of water molecules per $-\text{SO}_3\text{H}$ group; \bar{V}_M , the partial molar volume of membrane; and \bar{V}_2 is the partial molar volume of water, $\bar{V}_2 = 18 \text{ cm}^3 \text{ mol}^{-1}$.

$$\bar{V}_M \approx \frac{\text{EW}}{\rho_0}$$

where EW is membrane equivalent weight, for Nafion 117, 115, 1135 and 112, it is 1100; ρ_0 is density of dry membrane, $\rho_0 = 2.05 \text{ g cm}^{-3}$.

$$\lambda_1^0 = \lambda_{1,298}^0 \exp \left[-\frac{E_\eta}{R} \left(\frac{1}{T} - \frac{1}{298} \right) \right]$$

where $\lambda_{1,298}^0$ is equivalent conductance for H_3O^+ at infinite dilution at 298 K, $\lambda_{1,298}^0 = 349.8 \text{ S cm}^{-2} \text{ eq}^{-1}$. E_η is activation energy for viscosity of water, $E_\eta = 14 \text{ kJ mol}^{-1}$. R is universal gas constant, $R = 8.3143 \text{ J mol}^{-1} \text{ K}$. T is the operating temperature.

$$\alpha = \frac{(\lambda + 1) - \sqrt{(\lambda + 1)^2 - 4\lambda(1 - 1/K_{A,C})}}{2(1 - 1/K_{A,C})}$$

where $K_{A,C}$ is equilibrium constant for proton solvation in terms of concentrations.

$$K_{A,C} = K_{A,C,298} \exp \left[-\frac{\Delta H^\circ}{R} \left(\frac{1}{T} - \frac{1}{298} \right) \right]$$

where $K_{A,C,298}$ is equilibrium constant for proton solvation in terms of concentrations at 298 K, $K_{A,C,298} = 6.2$. ΔH° is the enthalpy change, $\Delta H^\circ = -52.3 \text{ kJ mol}^{-1}$.

The critical exponent q is universal constant predicted to be about 1.5. The threshold value ε_0 is best determined from experiments as a fitting parameter with value of 0.06. δ , a value of 5.5, is judiciously adjusted according to the experiment.

Fig. 3 shows the relationship between the conductivity of the membrane and the water loading (the number of water molecules

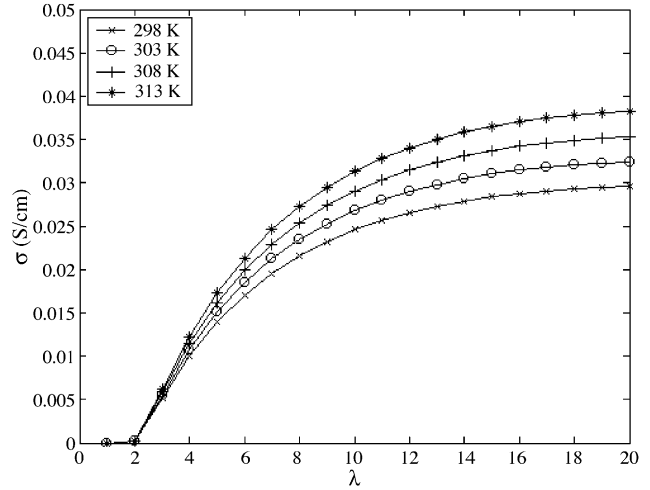


Fig. 3. Relationship between conductivity of the membrane and water loading.

per $-\text{SO}_3\text{H}$ group) under different operating temperature. It is noted that when the water loading is below five, the conductivity drops significantly. For a fixed water loading, the conductivity improves with the increase of temperature. The effect of temperature on conductivity diminished when the water loading drops toward the threshold value of two.

5. Relationship between water loading and relative humidity

Membrane water content/loading (λ) is essentially the ratio of the number of water molecules per SO_3^-H^+ charge site. It can be determined from the activity of water vapour at the electrode/membrane interface. The activity (a_2) in the vapour phase is defined as:

$$a_2 = \phi = \frac{p_w}{p^{\text{sat}}}$$

where ϕ is the relative humidity, p_w and p^{sat} are the pressure and saturation pressure of the water vapour, respectively. The latter is fitted to the following empirical expression using the thermodynamic data of the water:

$$\log_{10} p^{\text{sat}} = -2.1794 + 0.02953T - 9.1837 \times 10^{-5} T^2 + 1.4454 \times 10^{-7} T^3$$

Zawodzinski et al. [14] measured and expressed the water loading as a function of water activity for the Nafion 117 membrane. Fig. 4 in ref. [14] shows the experimental fit of “isopiestic curve” measured at 30 °C. This experimental fit of λ versus a can be expressed by the following equation.

$$\lambda_{(30\text{C})} = 0.043 + 17.81a_2 - 39.85a_2^2 + 36a_2^3 \quad \text{for } 0 \leq a_2 \leq 1$$

When $a_2 = 1$, the value of λ in equilibrium with saturated water vapour is 14 (or more specifically, 14.003) H_2O molecules per charged site. When the membrane, following partial drying, is immersed in liquid water at 80 °C, λ can go as high as 16.8. The λ is assumed to increase linearly from 14 to 16.8 when the mole

fraction of water exceeds the saturation from $x_{w,sat}$ to $3x_{w,sat}$, i.e.,

$$\lambda = 14 + 1.4(a_2 - 1) \quad \text{for } 1 \leq a_2 \leq 3$$

Alternatively, the water sorption characteristics of Nafion can be well modelled by Brunauer–Emmett–Teller (BET) equation [15]:

$$\frac{\lambda}{\lambda_m} = \frac{[Ca_2/(1 - a_2)][1 - (n_2 + 1)a_2^{n_2+1} + n_2a_2^{n_2+1}]}{1 + (C - 1)a_2 - Ca_2^{n_2+1}}$$

where λ_m is the water loading at monolayer coverage, C is BET constant, a_2 is the activity, or relative humidity, and n_2 is total number of water layers sorbed on the pore surface.

The monolayer coverage λ_m was estimated from knowledge of the specific pore surface area S by:

$$\lambda_m = \frac{S \text{ EW}}{\rho_0 N_A A_2}$$

where the surface area occupied by an adsorbate molecule on the pore surface and can be estimated from:

$$A_2 = 1.091 \left(\frac{MW_2}{\rho_2 N_A} \right)^{2/3}$$

For Nafion, S is $210 \text{ m}^2 \text{ cm}^{-3}$. N_A is the Avogadro’s constant; MW_2 , the molecular mass of water; and ρ_2 is the density of water. C and n_2 are treated as fitting parameters, $C = 150$ and $n_2 = 13.5$.

Fig. 4 shows the adsorption isotherm for water loading by Nafion equilibrated with water vapour. If water loading of five is required, the relative humidity should be over 60%. Combining the relationship between conductivity and water loading and the relationship between water loading and relative humidity, the relationship between conductivity and relative humidity can be obtained, which is shown in Fig. 5.

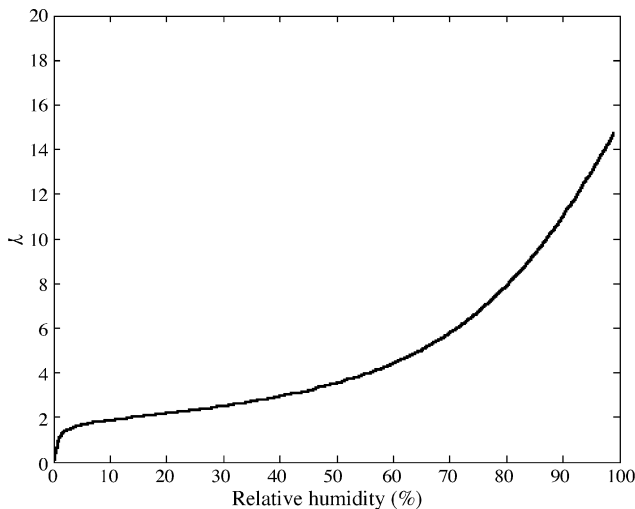


Fig. 4. Adsorption isotherm for water loading by Nafion from water vapor.

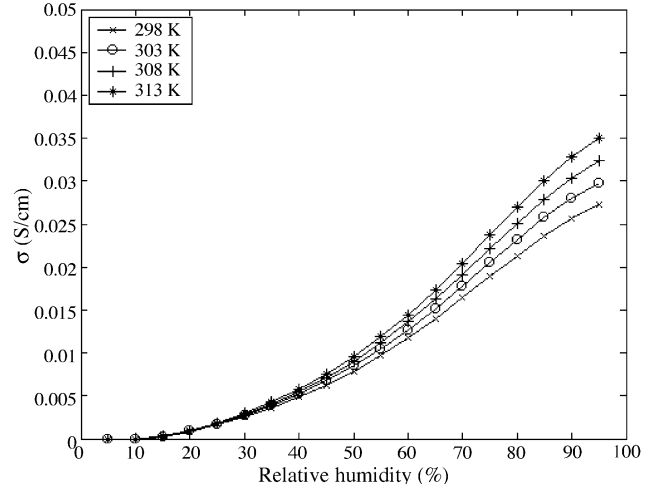
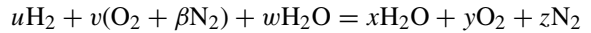


Fig. 5. Relationship between conductivity of membrane and relative humidity.

6. Relationship between relative humidity and stoichiometric number of air

The water activity is contributed from both the wet air and water arising from the electrochemical reaction of $\text{H}_2\text{--O}_2$. If the entire system is taken to be held at a constant temperature, and the water vapour is assumed to be an ideal gas and well mixed in the cathodic chamber, then the partial pressure of the water can be calculated from the known conditions of the air (relative humidity and temperature of the atmospheric air) and the operating air stoichiometric number.

The overall reaction can be expressed as:



In the equation,

$$u = 1, \quad v = \frac{\text{Sto } p_t}{2(p_t - \phi_{in} p_{in}^{sat})}, \quad w = \frac{\text{Sto } \phi_{in} p_{in}^{sat}}{2(p_t - \phi_{in} p_{in}^{sat})}$$

and

$$x = 1 + \frac{\text{Sto } \phi_{in} p_{in}^{sat}}{2(p_t - \phi_{in} p_{in}^{sat})}, \quad y = \frac{\text{Sto } p_t}{2(p_t - \phi_{in} p_{in}^{sat})} - \frac{1}{2},$$

$$z = \frac{\beta \text{Sto } p_t}{2(p_t - \phi_{in} p_{in}^{sat})}$$

where Sto is the stoichiometric number of air; p_t the total pressure; ϕ_{in} the relative humidity of inlet air, which is assumed to be 70% in this study; p_{in}^{sat} , the saturated vapour pressure evaluated at 25°C ; and $\beta = 3.762$, which is the volume ratio of nitrogen to oxygen in the air.

Thus, water partial pressure is:

$$\frac{p_w}{p_t} = \frac{2(p_t - \phi_{in} p_{in}^{sat}) + \text{Sto } \phi_{in} p_{in}^{sat}}{p_t + (1 + \beta)\text{Sto } p_t + \phi_{in} p_{in}^{sat}(\text{Sto} - 1)}$$

Fig. 6 shows the relationship between stoichiometric number of the air and relative humidity. With the same stoichiometric number, the higher the operating temperature, the lower the relative humidity will be. If the stoichiometric number is less than

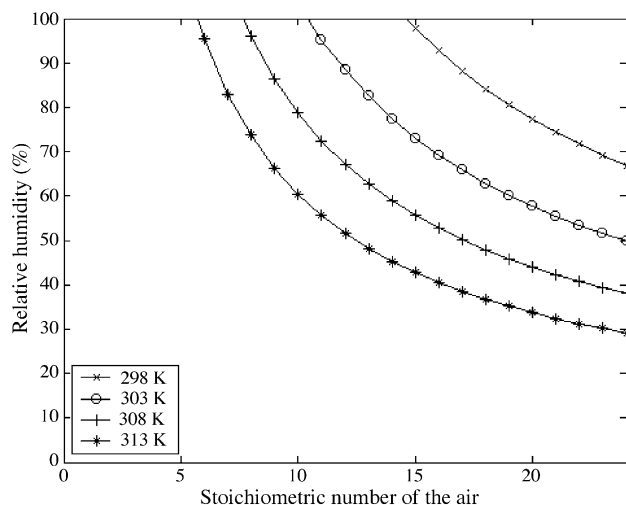


Fig. 6. Relationship between stoichiometric number of the air and relative humidity.

a critical value (i.e., 14.7, 10.5, 7.7, and 5.8 for temperature of 25, 30, 35 and 40 °C, respectively), the relative humidity can always reach 100%.

7. Relationship between conductivity and stoichiometric number of air

Finally, the relationship between conductivity of the membrane and stoichiometric number of the air is established, which is shown in Figs. 7 and 8. In Fig. 7, one can see that when the air stoichiometric number is low, the conductivity will remain the same over a range of air stoichiometric number because the membrane is wet under the relative humidity of 100%. The results show that when the stoichiometric number is increased above a certain threshold, the conductivity decreases. It is also shown that the higher the operating temperature, the higher the decreasing rate of conductivity will be. In Fig. 8, in the temperature range of 298–313 K, one can see that at low air stoichiometric number, the higher the operating temperature, the

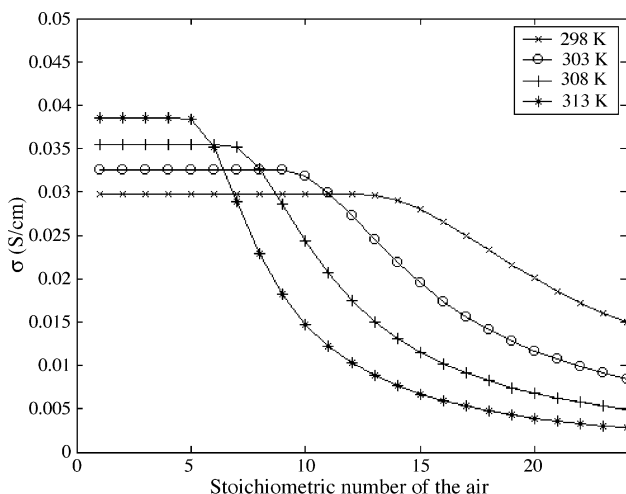


Fig. 7. Relationship between conductivity of membrane and stoichiometric number of the air.

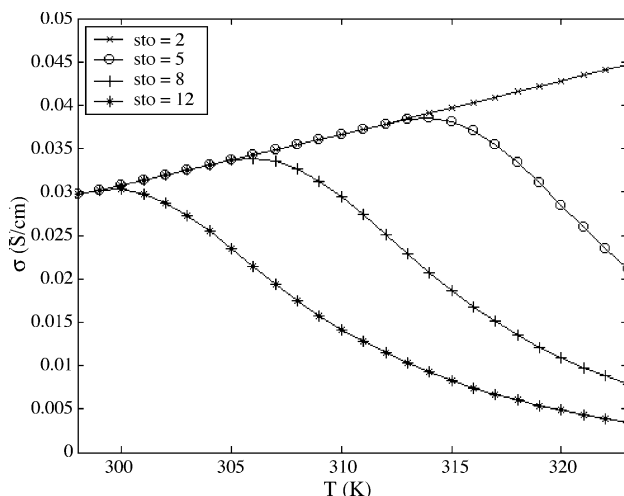


Fig. 8. Relationship between conductivity of membrane and operating temperature.

higher the conductivity will be; in the middle air stoichiometric number range, the conductivity increases with temperature until reaching a peak then decreases; at high air stoichiometric number, the peak conductivity is vague and the conductivity mostly decreases with the increase of operating temperature.

8. Conclusions

With the objective of maximized energy efficiency, the relationships between conductivity and water loading, water loading and relative humidity, and relative humidity and air stoichiometric number under different temperatures were established. Through these relationships, the conductivity, which is performance related, is correlated with the air stoichiometric number. Thus, the latter has become the sole control parameter for the small PEMFC stack operates at a fixed temperature under the above mentioned constraints. The results showed that, in the temperature range of 298–313 K, at low air stoichiometric number, the higher the operating temperature, the higher the conductivity will be; in the middle air stoichiometric number range, the conductivity increases with temperature until reaching a peak then decreases; at high air stoichiometric number, the peak conductivity is vague and the conductivity mostly decreases with the increase of operating temperature. For a fixed operating temperature, the conductivity drops dramatically when the air stoichiometric number exceeds the threshold value. The higher the operating temperature, the more rapid drop of the conductivity against the increased air stoichiometric number will be.

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References

- [1] S.H. Chan, H.K. Ho, O.L. Ding, Analysis of a simple solid oxide fuel cell system with gas dynamic in afterburner and connecting pipes, *Fuel Cells* 5 (1) (2005) 25–33.
- [2] J.H. Lee, T.R. Lalk, Modeling fuel cell stack systems, *J. Power Sources* 73 (1998) 229–241.
- [3] R.F. Mann, J.C. Amphlett, M.A.I. Hooper, H.M. Jensen, B.A. Peppley, P.R. Roberge, Development and application of a generalized steady-state electrochemical model for a PEM fuel cell, *J. Power Sources* 86 (2000) 173–180.
- [4] R. Jiang, D. Chu, Stack design and performance of polymer electrolyte membrane fuel cells, *J. Power Sources* 93 (2001) 25–31.
- [5] S.H. Chan, W.A. Tun, Catalyst layer models for proton exchange membrane fuel cells, *Chem. Eng. Technol.* 24 (1) (2001) 51–57.
- [6] N. Rajalakshmi, M. Raja, K.S. Dhathathreyan, Evaluation of current distribution in a proton exchange membrane fuel cell by segmented cell approach, *J. Power Sources* 112 (2002) 331–336.
- [7] A. Kumar, R.G. Reddy, Effect of channel dimensions and shape in the flow-field distributor on the performance of polymer electrolyte membrane fuel cells, *J. Power Sources* 113 (2003) 11–18.
- [8] S.H. Chan, S.K. Goh, S.P. Jiang, A mathematical model of polymer electrolyte fuel cell with anode CO kinetics, *Electrochim. Acta* 48 (2003) 1905–1919.
- [9] D.M. Bernardi, M.W. Verbrugge, Mathematical model of a gas diffusion electrode bonded to a polymer electrolyte, *AIChE J.* 37 (8) (1991) 1151.
- [10] T.E. Springer, T. Rockward, T.A. Zawodzinski, S. Gottesfeld, Model for polymer electrolyte fuel cell operation on reformat feed—effects of CO, H₂ dilution, and high fuel utilization, *J. Electrochem. Soc.* 148 (1) (2001) A11.
- [11] T. Hottinen, M. Mikkola, P. Lund, Evaluation of planar free-breathing polymer electrolyte membrane fuel cell design, *J. Power Sources* 129 (2004) 68–72.
- [12] B. Carnes, N. Djilali, Systematic parameter estimation for PEM fuel cell models, *J. Power Sources* 144 (2005) 83–93.
- [13] T. Thampan, S. Malhotra, H. Tang, R. Datta, Modeling of conductive transport in proton-exchange membranes for fuel cells, *J. Electrochem. Soc.* 147 (9) (2000) 3242–3250.
- [14] T.A. Zawodzinski, C. Derouin, S. Radzinski, R.J. Sherman, V.T. Smith, T.E. Springer, S. Gottesfeld, Water uptake by and transport through Nafion 117 membranes, *J. Electrochem. Soc.* 140 (1993) 1041–1047.
- [15] A.W. Adamson, A.P. Gast, *Physical Chemistry of Surfaces*, sixth ed., Wiley Interscience, New York, 1997, p. 622.