

CHAPTER 2 THEORIES AND LITERATURE REVIEWS

The theories and literature reviews concerning to the proton exchange membrane fuel cell and passivity concept will be presented in this chapter.

2.1 Theories

2.1.1 The proton exchange membrane fuel cell (PEMFC)

Proton exchange membrane fuel cell, also known as polymer exchange membrane fuel cell (PEMFC) is one of the most promising sources of renewable energy. They can be considered as green power because they are environmental clean.

PEMFC is an electro-chemical device that converts the chemical energy of hydrogen and oxygen to electrical power and heat. It consists of negatively charge electrode (anode), positively charge electrode (cathode), and an electrolyte membrane. Figure 2.1, hydrogen is fed into the anode side while air is fed into the cathode side. Hydrogen is diffused to the anode diffusion layer to active catalyst layer, where hydrogen generates free hydrogen protons and electrons. Then, the hydrogen protons transport from the anode to the cathode through the electrolyte membrane, and the electrons are carried out to the cathode over the external circuit. In meantime, oxygen which enters to the cathode channel diffuses through the cathode diffusion layer to active catalyst, where the oxygen dissociates and combines with protons and electrons to form water, while heat and current are generated[3]. A typical electro-chemical reaction of PEMFC is following:

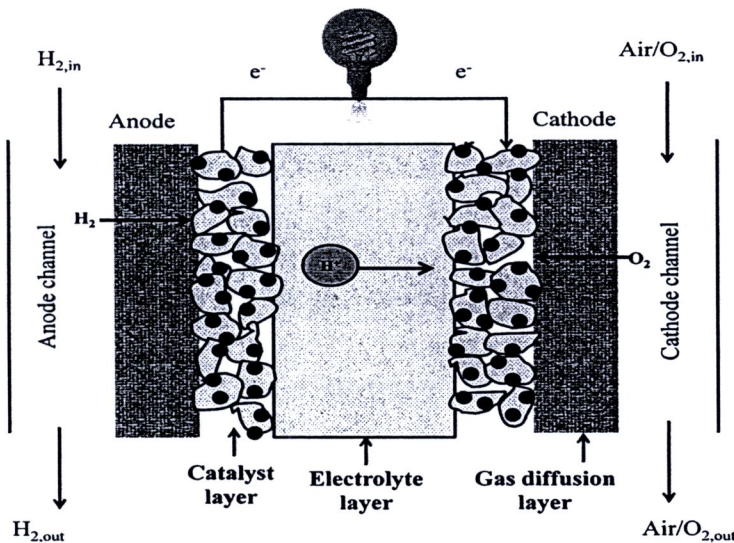
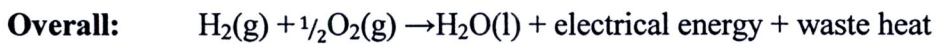
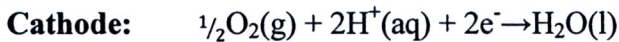
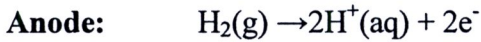


Figure 2.1 Schematic illustration of proton exchange membrane (PEMFC)

In practice, a 5-kW fuel cell stack, such as a Ballard MK5-E PEMFC stack, is widely used not only for an experiment but also for a practicable. On the cathode side, an air supply system containing an air flow controller is required to maintain the oxygen partial pressure. The humidifier is required to prevent dehydration or flooding of fuel cell [5]. In addition, a heat exchanger, a water tank, and a pump may be needed for water and heat management in the fuel cell systems.

In terms of the electricity produced by the electrochemical process, PEMFC is similar to a battery but it requires continuous reactants supply to generate constant electricity, while a battery does not need any external reactants supply except for recharging when external electricity is applied [6].

In 1960, the first practical PEM fuel cell was developed for using with space applications. Since then, fuel cell has been successful, it was continued for applying with terrestrial applications, just the last 10 to 15 years have been remarkable for terrestrial applications. In 1993, Ballard power system demonstrated fuel cell powered buses. All major automotive manufacturers developed prototypes of fuel cell vehicle in the late 1990s and the early 2000s.

For stationary power applications, more than 2500 fuel cell stationary power systems have been installed globally for hospital, building, utility power plants, and so on. In 2005, Samsung Electronics also unveiled a prototype of fuel cells that run a laptop for 15 hours for portable power applications. However, there are many challenges and technical issues of commercialization of fuel cells. The most significant problems are reducing fuel cells costs as well as improving fuel cell operating liability.

2.1.1.1 PEMFC voltage model

Fuel cell voltage indicates the performance of fuel cell. Under normal operation, a simple fuel cell typically produces 0.5-0.9 V based on the polarization I-V curve, which expresses the relationship between the stack voltage and the load current [5]. To produce a higher voltage, multiple cells are connected in series. Figure 2.2 shows that their relationship is nonlinear and mainly depends on current, stack temperature, reactant partial pressure, and membrane humidity [5, 7]. The output voltage of single cell can be defined as following [8]:

$$V_{fc} = E_{Nernst} + \eta_{act} + \eta_{ohm} \quad (2.1)$$

In this expression, E_{Nernst} is the thermodynamic potential of the cell (V) and represents its reversible voltage:

$$E_{Nernst} = 1.229 - (8.5 \times 10^{-4})(T - 298.15) + (4.308 \times 10^{-5})T[\ln(P_{H_2}) + \frac{1}{2}\ln(P_{O_2})] \quad (2.2)$$

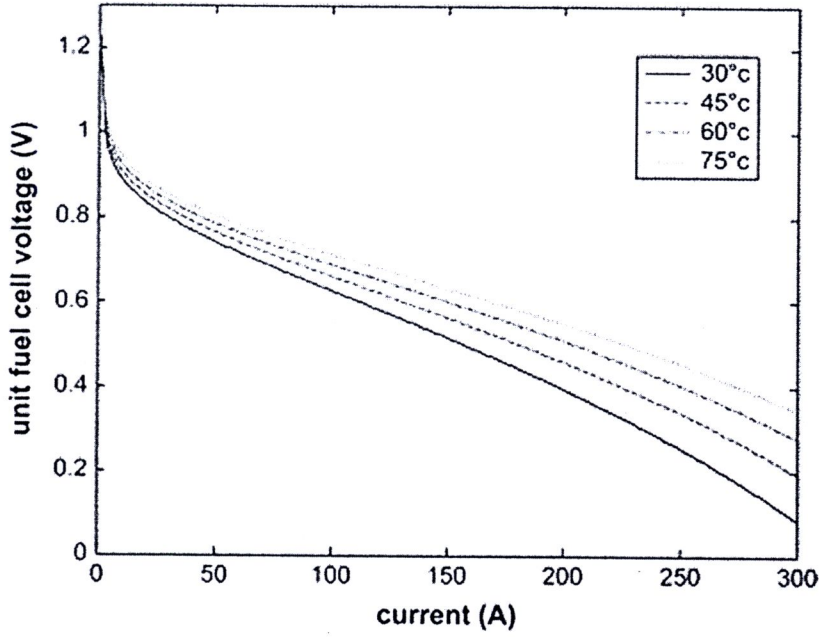


Figure 2.2 Polarization V-I curve (Ballard Mark V PEMFC) [9]

η_{act} is the voltage drop (V) due to the activation of the anode and the cathode [10]:

$$\eta_{act} = \xi_1 + (\xi_2 T) + (\xi_3 T \ln I) + (\xi_4 T \ln C_{O_2}) \quad (2.3)$$

The oxygen concentration showed in equation 2.3 can be calculated using the Henry's law as the following equation:

$$C_{O_2} = \frac{P_{O_2}}{(5.08 \cdot 10^{-6}) \exp\left(\frac{-498}{T}\right)} \quad (2.4)$$

η_{ohm} is ohmic voltage drop, a measure of ohmic voltage drop associated with the conduction of the proton through the solid electrolyte and electrons through the internal electronic resistances.

$$\eta_{ohm} = -iR^{internal} \quad (2.5)$$

$$R^{internal} = \xi_5 + \xi_6 T + \xi_7 I \quad (2.6)$$

These equations are combined in an appropriate algorithm, and are able to predict the voltage output of proton exchange membrane fuel cell (PEMFC) [11].

2.1.1.2 PEMFC thermal model

The electric heat produced from the electrochemical reaction is one of the important problem for PEMFC. When PEMFC is operated, the stack temperature is getting high and may be cause of membrane dehydration. PEMFC will give low performance due to the proton transportation in membrane becomes difficult. To maintain the performance of PEMFC, the stack temperature should be controlled to the optimal level [6]. Figure 2.2 shows that the polarization curve is shifted upward as the temperature increases.

Although the temperature control is vital for the fuel cell operation, a quite number of control oriented PEMFC models [10, 12-13] have been developed under the assumption that the operating temperature is constant due to the complexity and long time constant (over 2000 sec) of temperature [6]. In this study, the temperature is defined as one of the state variables and a control strategy is developed for study the transient thermal model of PEMFC. The energy balance of thermal PEMFC thermal model is described as follow [6]:

$$\dot{Q}_{stack} = C_t \frac{dT_s}{dt} = \dot{P}_{tot} - \dot{P}_{elec} - \dot{Q}_{cool} - \dot{Q}_{loss} \quad (2.7)$$

The total power (\dot{P}_{tot}) is delivered into the stack directly related to the amount of hydrogen consumed,

$$\dot{P}_{tot} = \dot{m}_{H_2,used} \Delta H = \frac{NI}{2F} \Delta H \quad (2.8)$$

The electrical power output is given by:

$$\dot{P}_{elec} = V_{stack} I_{stack} \quad (2.9)$$

The rate of heat removal by the cooling water is directly related to the cooling water flow in the heat exchanger. The relationship is as follows [14]:

$$\dot{Q}_{cool} = \dot{m}_{cw} c_{p,cw} (T_s - T_c) \quad (2.10)$$

The heat loss by the surface of the stack is calculated as follow:

$$\dot{Q}_{loss} = hA_{stack} (T_s - T_{amb}) \quad (2.11)$$

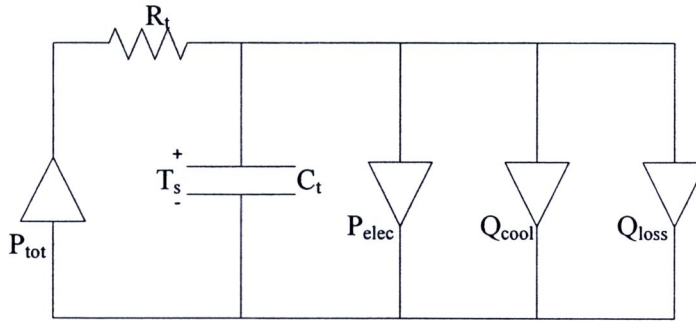


Figure 2.3 The simple thermal circuit of PEMFC system

Figure 2.3 shows the simple thermal circuit of PEMFC system which four dependent energy sources from Equation 2.7 are involved. The total power (\dot{P}_{tot}) depends on the hydrogen consumption based on the load changes and the electrical power (\dot{P}_{elec}) depends on the load current. The cooling power (\dot{Q}_{cool}) and the heat loss by the surface of the stack (\dot{Q}_{loss}) are a function of the fuel cell stack temperature changes. In Figure

of the stack (\dot{Q}_{loss}) are a function of the fuel cell stack temperature changes. In Figure 2.3, the various load changes cause the changes in PEMFC stack temperature, therefore the PEMFC voltage (V_{stack}) and the current (I) can be used as the external input or disturbance variables, while the stack temperature is the output variable of PEMFC system.

2.1.1.3 PEMFC mass model

The transient dynamic in anode and cathode channels are also considered in this work by applying the dynamic mass principle on its respective control volumes. In the anode channel modeling, it is simplified by assuming pure hydrogen flows into the channel. Further, partial hydrogen diffuses through anode electrode into the anode active layer where it is consumed by an electrochemical reaction. The dynamic model of the anode channel is obtained by employing mass conservation law on the anode control volume as follow [9]:

$$\frac{V_{an}}{RT_s} \frac{dP_{H_2}}{dt} = \dot{m}_{H_2,in} - \dot{m}_{H_2,out} - \dot{m}_{H_2,diff} \quad (2.12)$$

The excess hydrogen is re-circulated to the anode and the throttle is assisted to regulate the inlet flowrate of hydrogen, thereby the hydrogen outlet flowrate ($\dot{m}_{H_2,out}$) is assumed to,

$$\dot{m}_{H_2,out} = k_{an}(P_{H_2} - P_{atm}) \quad (2.13)$$

The term of $\dot{m}_{H_2,diff}$ is the hydrogen consumed by the electrochemical reaction depends on the load current, which can be described as follow:

$$\dot{m}_{H_2,diff} = \frac{NI}{2F} \quad (2.14)$$

The cathode control volume analysis is similar to the anode control volume, which pure oxygen flows into the channel. The dynamic model of the cathode channel is described below [9]:

$$\frac{V_{cat}}{RT} \frac{dP_{O_2}}{dt} = \dot{m}_{O_2,in} - k_{cat}(P_{O_2} - P_{atm}) - \frac{NI}{4F} \quad (2.15)$$

As the by product from the electrochemical reaction is water, the relationship between hydrogen and oxygen convert to water is described by employing mass conservation principle on electrochemical reaction [2].

$$\dot{m}_{H_2O,gen} = \dot{m}_{H_2,diff} + \dot{m}_{O_2,diff} \quad (2.16)$$

Regarding the water product of the electrochemical reaction, flooding phenomenon is also considered in this work. Flooding occurs as the relative humidity (RH) in the

membrane [15]. Then, the mass transfer will decrease gradually. The relative humidity can be calculated via Raoult's law as follow [16]:

$$RH(\%) = \frac{P_{H_2O}}{P_{H_2O}^{sat}} \times 100\% \quad (2.17)$$

$P_{H_2O}^{sat}$ is determined from Antoine's equation:

$$\log P_{H_2O}^{sat} = -2.1794 + 0.02953(T_s - 273) - 9.1873 \times 10^{-5}(T_s - 273)^2 + 1.4454 \times 10^{-7}(T_s - 273)^3 \quad (2.18)$$

This completes the modeling of PEMFC system.

2.1.2 Passivity and its relation properties

2.1.2.1 State space

The basic concepts of passive systems are introduced in this section. The concept of passivity is fundamentally connected with dissipative and is a special case of dissipative. Consider the definition of dissipative systems as following control affine system [17]:

$$\dot{x} = Ax + Bu + Ex_0 \quad (2.19)$$

$$y = Cx + Du \quad (2.20)$$

Equations 2.19-20 are represented in the form of standard state-space concept which is normally used in the modern control system. The feature of the state-space approach that sets it apart from the frequency-domain approach is the representation of the processes under examination by systems of first-order differential equations [17]. Since the system of interest has the standard state-space, the transfer function (matrix) of the system and disturbance are given explicitly by

$$G_p(s) = C(sI - A)^{-1}B + D \quad (2.21)$$

$$G_d(s) = C(sI - A)^{-1}E \quad (2.22)$$

2.1.2.2 Passivity concept

Dealing with passivity concept, the system has $n \times n$ transfer function matrix in the form of Equation 2.21 is said to be passive, or positive real (PR) system if they stay in all these three conditions [18]:

- (1) $G(s)$ is analytical in $\text{Re} > 0$
- (2) $G(j\omega) + G^*(j\omega) \geq 0$ for all frequency ω that $j\omega$ is not a pole of $G(s)$
- (3) If there are poles p_1, p_2, \dots, p_m of $G(s)$ on the imaginary axis, they are non-repeated and the residue matrices at the poles $\lim_{s \rightarrow p_i} (s - p_i) G(s)$ are Hermitian and positive semi-definite.

In addition, the system is said to be strictly passive or strictly positive real (SPR) if the above two conditions are changed to:

- (1) $G(s)$ is analytical in $\text{Re} \geq 0$
- (2) $G(j\omega) + G^*(j\omega) > 0$ for $\omega \in (-\infty, +\infty)$

To analyze the stability of the linear system, the passivity concept state that the feedback system comprised of a passive system and a strictly passive system is asymptotically stable. This concept is used for the multi-loop control system as shown in Figure 2.4.

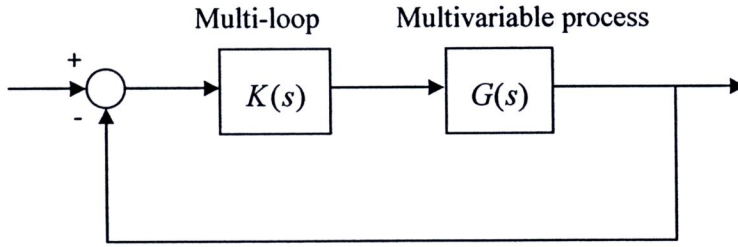


Figure 2.4 Multi-loop control system [4]

If the multi-variable process system $G(s)$ is strictly passive, then the closed-loop system will be stable if the multi-loop controller $K(s)$ is passive.

To facilitate future stability analysis, defining $G^+(s) = G(s)U$, where U is a diagonal matrix with either 1 or -1 along the diagonal. The signs of U elements are determined such that the diagonal elements of $G^+(s)$ are positive at steady state, that is $G_{ij}^+(0) \geq 0$, $i = 1, \dots, n$, it can be denoted that,

$$K^+(s) = U^{-1}K(s) = UK(s) \quad (2.23)$$

$$G^+(s) = G(s)U \quad (2.24)$$

To analyze whatever the system is passive or not, the system can be verified easily by using passivity index (ν_F) from the passivity concept.

2.1.2.3 Passivity Index (ν_F)

Passivity index $\nu_F[G, \omega]$ indicates how far the system $G(s)$ is being passive and it is negative if the system is passive [18]. The passivity index can be defined as following:

$$\nu_F(G, \omega) = -\lambda_{\min} \left(\frac{1}{2} [G(j\omega) + G^*(j\omega)] \right) \quad (2.25)$$

However, if the system is non-passive with positive value of passivity index, a stable and minimum phase transfer function $w(s)$ is required to add into the system. The

passivity concept states that if $w(s)$ is chosen such that Equation 2.28, then $H(s)$ is strictly passive.

$$\nu(w(s), \omega) < -\nu(G(s), \omega) \quad (2.26)$$

$$H(s) = G(s)U + w_p(s)I \quad (2.27)$$

I is the identity matrix of size n which is the $n \times n$ square matrix with ones on the main diagonal and zeros elsewhere.

For the system $G^+(s) = G(s)U$, is positive real at steady state, then $w_p(s)$ can be chosen to have the following form,

$$w_p(s) = \frac{ks(s+a)}{(s+b)(s+c)} \quad (2.28)$$

Where a, b, c and k are positive and real parameters to be determined by solving the following optimization problem 1 obtained from the passivity concept [18].

Problem 1:
$$\min_{a,b,c,k} \sum_{i=1}^m (\operatorname{Re}(w_p(j\omega_i)) - \nu_s(G^+(s), \omega_i))^2$$

subject to $\operatorname{Re}(w_p(j\omega_i)) > \nu_s(G^+(s), \omega_i) \quad \forall i = 1 \dots m \quad (2.29)$

2.1.2.4 Passive controller design

For the passive system, the controller achieves the following condition will be the passive controller with decentralized unconditional stability.

$$1. \operatorname{Re} \left\{ \frac{k_i^+(j\omega)}{1 - \nu_s(G(s), \omega) k_i^+(j\omega)} \right\} \geq 0 \quad \forall \omega \in R, i = 1, \dots, n \quad (2.30)$$

$$2. K(s) \text{ is analytic in } \operatorname{Re}(s) > 0 \quad (2.31)$$

Where $k_i^+(s) = U_{ii} k_i(s)$. The (i, i) th element in matrix U indicates whether the corresponding i th loop is direct acting or reverses acting. The decentralized unconditional stability condition given in Equations 2.30-31 imply:

$$1. k_i^+(s) \text{ is passive and} \quad (2.32)$$

$$2. \left| k_i^+(j\omega) - \frac{1}{2\nu_s(G(s), \omega)} \right| \leq \left| \frac{1}{2\nu_s(G(s), \omega)} \right| \quad \forall \omega \in R, i = 1, \dots, n \quad (2.33)$$

At frequency ω , any $k_i^+(j\omega)$ which satisfies the above two conditions will be confined within the disk on the s plane, as shown in Figure 2.6. The amplitude ratio of the multi-loop controller at frequency ω is limited to $|1 / \nu_s(G(s), \omega)|$ which indicates the performance of the passivity-based decentralized unconditionally controllers.

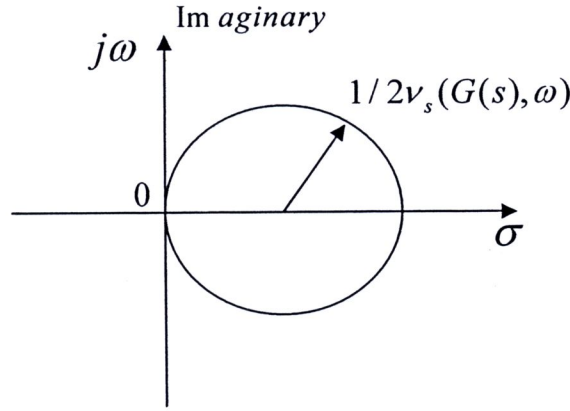


Figure 2.5 Passivity index and controller gain

For multi-loop control system, the multi-loop controllers can be designed by using the proposed stability condition in sections 2.1.2.2-3 in order to achieve the close-loop stability. The multi-loop PI controller synthesis from Equations 2.32-33 is converted into the following optimizations problem 2 to minimize the sensitivity function of each loop [18].

Problem 2:

$$\min_{k_{c,i}^+, \tau_{I,i}} (-\gamma_i)$$

$$\text{subject to} \quad \left| \frac{1}{1 + G_{ii}^+(j\omega)k_{c,i}^+[1 + 1/\tau_{I,i} \times j\omega]} \frac{\gamma_i}{j\omega} \right| < 1 \quad (2.34)$$

$$\text{and} \quad \tau_{I,i}^2 \geq \frac{k_{c,i}^+ v_s(\omega)}{[1 - k_{c,i}^+ v_s(\omega)]\omega^2} \quad \forall \omega \in R, i = 1, \dots, n \quad (2.35)$$

For the multi-loop control system as shown in Figure 2.4 comprising a stable subsystem $G(s)$ and a multi-loop controller $K(s) = \text{diag}\{k_i(s)\}$, $i = 1, \dots, n$, if a stable and minimum phase transfer function $w(s)$ is chosen such that Equation 2.28, then the closed-loop system will be stable. For any loop $i = 1, \dots, n$, the passive controllers are design as follow:

$$k_i'(s) = k_i^+(s)[1 - w(s)k_i^+(s)]^{-1} \quad (2.36)$$

where $k_i^+(s) = U_{ii}k_i(s)$ and $U = \text{diag}\{U_{ii}\}$, $i = 1, \dots, n$,

To analyze the stability of multi-loop control system, Figure 2.7 is extended from Figure 2.4. Where $K'(s) = \text{diag}\{k_i'(s)\}$ and $W(s) = w(s)I$.

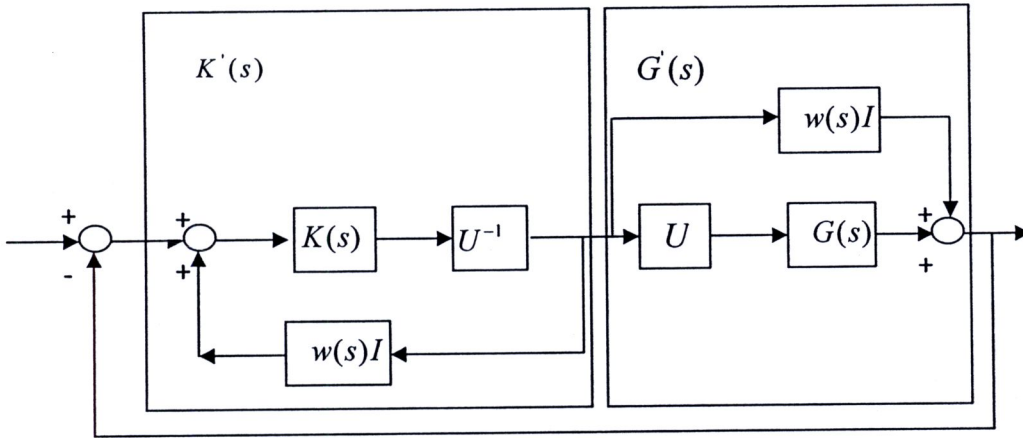


Figure 2.6 Passive system

From Figure 2.7, if $G'(s)$ is SPR and $K'(s)$ is PR, the closed-loop system will be stable. Because $K^+(s)$ is diagonal, so is the subsystem of $K'(s)$. Then, $K'(s)$ is passive if its diagonal element $k_i'(s)$ is passive for each loop $i = 1, \dots, n$. This completes the passive control system.

2.2 Literature reviews

Since the aim of this work is to apply the stability analysis based on passivity concept to control the proton exchange membrane fuel cell (PEMFC) by multi-loop passive controller, the publications related to this work are analyzed thoroughly.

To develop the control system of PEMFC, PEMFC has to be accurately modeled to apply the suitable nonlinear control technique. Unfortunately, many PEMFC models have been reported were mainly for experimental verification other than control design. Recently, Purkrushpan et al. [12] developed a 5 kW PEMFC Ballard stack model that included flow characteristics and dynamics of the manifold (anode and cathode), reactant partial pressures, and membrane humidity.

The PEMFC models proposed by Purkrushpan et al. [12] and L. Y. Chiu et al. [19] were linearized via a Taylor series expansion for prediction of the fuel cell phenomenon by analyzing an electrochemical reaction, the thermodynamics, and the fluid mechanics. However, because of the highly nonlinear PEMFC model, it is challenged to develop a nonlinear controller for control the PEMFC system [6].

To develop the PEMFC dynamic model, Xue et al. [3] proposed the model of mass and heat transfer in PEMFC by dividing the PEMFC into 3 control volumes which were the anode channel, the cathode channel, and the fuel cell body. The fuel cell body in their model consisted of anode and cathode gas diffusion layer, anode and cathode catalyst, and electrolyte membrane. This model was used to study the transient response of the



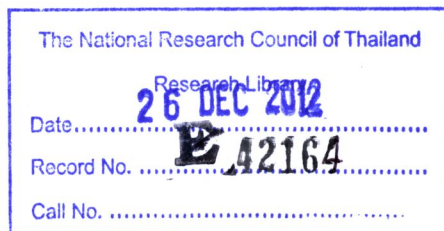
control volumes had similar trend which differ insignificantly. This clearly demonstrated the advantage of dividing the fuel cell system into three control volumes.

Due to the value product from PEMFC is DC electricity. Sharifi et al. [11] used the same modeling approach of Xue X. to study the effect of load current (I) to partial pressure of reactant and cell voltage using open-loop system. They also developed the mathematical model for PEMFC steady-state voltage to study the affect of the operating condition to the cell voltage. After step changed of load current from 10A to 50A, the consumption of hydrogen and oxygen for electrochemical reaction was getting high and mass of gases within the channel decreased. The cell voltage decreased immediately due to an increasing of ohmic and activation polarization. In conclusion, the load current density, the stack temperature, and the partial pressure of hydrogen and oxygen affected to the cell voltage of PEMFC.

To develop an advance PEMFC control system, Ahn et al. [20] studied the temperature effect on a stack, and also proposed the new temperature control strategy based on a thermal circuit. They proposed the proportional and integral (PI) controllers for the thermal circuit in order to minimize the temperature effect to PEMFC. Wu et al. [9] proposed the multi-loops feedback control design to control both stack temperature and oxygen excess ratio in the cathode at the optimal level. The controlled variables were specified independently by manipulation of air (oxygen) and water flowrates, respectively. Since the high current demand degraded the performance of fuel cell, a multi-loops feedback control scheme for PEMFC system was utilized under the current load changed. By the closed-loop simulation results, the manipulation of both air (oxygen) and water flowrates were verified to accurately dominate the operation and performance of PEMFC system.

However, the important problem of PEMFC is membrane damage from flooding or dehydration due to the electric heat from the electrochemical reaction. Therefore, the control strategy for membrane humidity is proposed in this work.

Zhang et al. [18] presented an approach to decentralized (multi-loop) failure-tolerant control design using the passivity concept. In their work, the passivity index was used to measure how far a system of concern was from being passive. A diagonally scaling technique was developed to reduce the conservativeness of passivity index. Then, weighting function, a minimum phase and stable scalar transfer function was added to obtain the passive system. The decentralized passivity-based PI controller was designed to control each loop individually. They studied the response of distillation system with 4×4 transfer function metric namely the step changed response of each loop control, the step changed in all loops and one loop failed. The result showed that the proposed control achieved good performance in all cases. This was because the passivity index reflected the overall stability both the interactions and dynamics of the entire process system.



Bao et al. [21] provided a new approach to stability analysis for multi-loop control systems based on passivity theorem. The destabilizing effect of interactions of a multi-input multi-output system was studied using the passivity index. The decentralized unconditional stability condition which implied closed-loop stability of decentralized control systems under control loop failure was derived. An easy-to-use stability test was performed on open-loop stable process transfer matrices, independent of controller design.

Multi-loop or decentralized control was the most widely used strategy in industrial process control. The selection of input and output variable pairing was always the first concern in designing such control systems. Bao et al. [21] proposed a new pairing method for multi-loop control for multivariable processes. Based on this passivity concept, the new pairing rule did not imply general diagonal dominance. A frequency-dependent passivity index was introduced to characterize the total destabilizing effect of both loop interactions and process dynamics. By using the passivity index, pairing schemes that provide best achievable control performance under decentralized control were chosen.

By the literature review, the multi-loop control of PEMFC system concerning to the stack temperature, the relative humidity and the cell voltage under the current load changed have been identified based on the passivity concept.