



# Control of a heat-integrated proton exchange membrane fuel cell system with methanol reforming

Wei Wu\*, Che-Chuan Pai

Department of Chemical and Materials Engineering, National Yunlin University of Science and Technology, Douliou, Yunlin 64002, Taiwan, ROC

## ARTICLE INFO

### Article history:

Received 6 March 2009

Accepted 18 May 2009

Available online 23 May 2009

### Keywords:

Fuel processing

Proton exchange membrane fuel cell

Heat integration

Multi-loop control

## ABSTRACT

This work presents a novel heat-integrated fuel cell stack system with methanol reforming. Its configuration is composed of fuel processing units (FPUs), proton exchange membrane (PEM) fuel cell stack, and heat exchangers (HEXs). Well mixed methanol and oxygen flows in contact with countercurrent flowing water dominates the production of hydrogen at the exit of FPUs and influences the stack temperature. The heat exchange connections can enhance the utilization of energy of FPUs. To ensure the stable steady-state operation, the model-free fuzzy incremental control scheme within the multi-loop feedback control framework is developed. Finally, the proposed system integration and control configuration are verified by closed-loop simulations.

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## 1. Introduction

Fuel cells are widely recognized as one of the most promising clean energy technologies of the future. The proton exchange membrane (PEM) fuel cell is quite suitable for residential or automotive applications [1], for the following reasons. (i) It can operate at a relatively low temperature; (ii) it has relatively high power density; (iii) its maintenance is simple. However, the efficiency of the PEMFC is strongly affected by the unsteady hydrogen feed flow, cell temperature, membrane dehydration and fast variations in load. Developing a source of pure hydrogen is a commercial challenge. Liquid hydrocarbons such as methanol, ethanol, and gasoline are usually treated as alternative hydrogen-rich fuel streams [2]. Notably, the methanol-fuelled reformer has been widely studied for fuel cell applications.

Toward the development of a fuel reforming system, Lindstrom and Pettersson [3] presented a compact methanol reformer for fuel cell vehicles; Lattner and Harold [4] developed a kinetic reactor model for autothermal reforming of methanol; Stamps and Gatzke [5] modeled the packed-bed methanol reformer; Choi and Stenger [6] proposed an integrated methanol reformer system to study the hydrogen yield and economic profit, and Wang and Wang [7] used a thermodynamic and exergetic analysis of a PEMFC stack system with methanol reforming. Recently, Xu et al. [8] explored the energy efficiency of a methane reforming system with heat integration. In fact, the methane-fuelled reforming system has been inadequately incorporated into the PEMFC system because of restrictions on

operating temperature and pollutant emissions. In our opinion, an integrated power generation system should depend on the synergy between heat integration and control design.

With respect to the fuel cell control problem, Lauzze and Chmielewski [9] provided a set of feedback structures, including power, temperature and relative humidity controllers; Woo and Benziger [10] showed that the proportional-integral-derivative (PID) control could accelerate the response of a PEMFC system to satisfy the load demand; Methekar et al. [11] adopted the multi-input multi-output (MIMO) PI control framework to control of the power density and temperature of a distributed parameter model of fuel cell systems, and Wu et al. [12] indicated that the MIMO control structure can ensure the highly efficient control performance when a low-order and simplistic model of the PEMFC systems is considered. However, the models-based predictive control schemes are adopted to improve the control performance by using the fuzzy Hammerstein model [13] or neural network techniques [14]. Moreover, fuzzy control schemes [15,16] and fuzzy neural networks [17] have been successfully applied to many fuel cell systems.

This article introduces the kinetics and modeling of fuel processing units (FPUs), including a methanol reformer (MR), heat exchangers (HEX), a water gas shift (WGS) reactor, and a preferential oxidation (PROX) reactor. In the proposed system configuration, the inlet methanol flow can influence the amount of hydrogen produced, and the countercurrent flow of water by virtue of heat exchange connections can improve energy utilization and regulate the stack temperature. Furthermore, the multi-loop fuzzy incremental control framework ensures the satisfactory tracking performance and reliability of the proposed heat-integrated system according to the closed-loop simulation.

\* Corresponding author.

E-mail address: [weiwu@yuntech.edu.tw](mailto:weiwu@yuntech.edu.tw) (W. Wu).

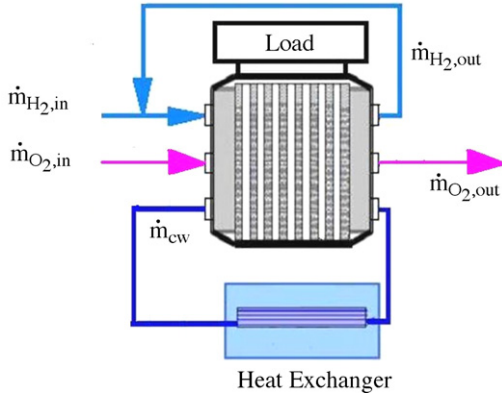


Fig. 1. Individual PEMFC stack system with cooling device.

## 2. PEMFC stack system

A Ballard 5 kW PEMFC stack system was constructed using the empirical-type model [18,19], which consists of 35 cells in series. Regarding the individual PEMFC stack system shown in Fig. 1, pure hydrogen is fed to the anode, excess hydrogen gas is re-circulated, and the stack temperature is changed by a heat exchange system with circulating water.

### 2.1. Anode and cathode flow models

Based on the assumption that all gases are ideal, the principles of mole conservation can be used to model the reactant flows at the anode and the cathode:

$$\begin{aligned} \frac{V_a}{RT} \frac{dP_{H_2}}{dt} &= \dot{n}_{H_2,in}^{FC} - k_a(P_{H_2} - P_{amb}) - \frac{nI}{2F} \\ \frac{V_c}{RT} \frac{dP_{O_2}}{dt} &= \dot{n}_{O_2,in}^{FC} - k_c(P_{O_2} - P_{amb}) - \frac{nI}{4F} \end{aligned} \quad (1)$$

where  $\dot{n}_{H_2,in}^{FC}$  and  $\dot{n}_{O_2,in}^{FC}$  are the hydrogen and oxygen inlet mole flow-rates, respectively. Notably, the inlet hydrogen flow-rate usually varies with the amount of hydrogen supplied, and the oxygen flow-rate can be determined by multiplying the air inlet flow-rate by the mole fraction of oxygen in air (21%).

### 2.2. Thermal model

The energy balance of the stack system is determined by the total power converted by the fuel  $H_2$  into electricity, the power consumed by the electrical load, the rate of heat removal by the coolant,  $\dot{Q}_{cool}$ , and the rate of heat loss at the stack surface,  $\dot{Q}_{loss}$ . The dynamic model is described by a couple of first-order differential equations:

$$\begin{aligned} C_t \frac{dT}{dt} &= \dot{E}_{tot} - \dot{E}_{elec} - \dot{Q}_{loss} - \dot{Q}_{cool} \\ \rho_w V_w C_{pw} \frac{dT_{c,out}}{dt} &= \dot{m}_{cw} C_{pw} (T_{c,in} - T_{c,out}) \\ &+ UA \left( T - \frac{T_{c,in} + T_{c,out}}{2} \right) \end{aligned} \quad (2)$$

where

$$\begin{aligned} \dot{E}_{tot} &= \frac{nI}{2F} \Delta H \\ \dot{E}_{elec} &= V_{stack} I \\ \dot{Q}_{loss} &= \frac{C_t (T - T_{amb})}{\tau} \\ \dot{Q}_{cool} &= (h_{cond} + h_{conv} I) \frac{(T - T_{c,in}) - (T - T_{c,out})}{\ln((T - T_{c,in}) / (T - T_{c,out}))} \end{aligned} \quad (3)$$

$T_{c,in}$  and  $T_{c,out}$  are the inlet and outlet water temperatures, respectively.  $\dot{m}_{cw}$  is the mass flow-rate of water;  $\rho_w$  is the density of water;  $V_w$  is the volume of the heat exchange system, and  $T$  is the stack temperature.

### 2.3. Polarization curve model

The polarization curve is generally used to specify the relation between the cell voltage  $V_{fc}$  and current density  $I$ . When a cell delivers power to the load, the load voltage  $E$  is reduced by the voltage drop, comprising of the activation overvoltage  $V_{act}$  and the ohmic overvoltage  $V_{ohm}$ . The output voltage of a single fuel cell is given by

$$V_{fc} = E - V_{act} - V_{ohm} \quad (4)$$

- (i) The open-circuit cell potential  $E$  is, as determined by the Nernst equation, given by

$$E = 1.229 - 8.5 \times 10^{-4} (T - 298.15) + \frac{RT}{2F} \ln [P_{H_2} (P_{O_2})^{0.5}] \quad (5)$$

- (ii) With respect to the dynamics of the activation overvoltage  $V_{act}$ , the first-order dynamic accounting for the effects of double layer capacitance charging at the electrode-electrolyte interfaces is described by

$$\frac{dV_{act}}{dt} = \frac{I}{C_{dl}} + \frac{E_{act}}{R_{act} C_{dl}} \quad (6)$$

where the activation resistance  $R_{act} = V_{act}/I$ ; the activation drop  $E_{act}$  is defined by

$$E_{act} = \beta_1 + \beta_2 T + \beta_3 T \ln(C_{O_2}) + \beta_4 T \ln(I), \quad (7)$$

and the parametric coefficients  $\beta_1, \dots, \beta_4$  are expressed as

$$\begin{aligned} \beta_1 &= -0.948 \\ \beta_2 &= 0.00286 + 0.0002 \ln(A_{fc}) + 4.3 \times 10^{-5} \ln(C_{H_2}) \\ \beta_3 &= 7.6 \times 10^{-5} \\ \beta_4 &= -1.93 \times 10^{-4} \end{aligned} \quad (8)$$

with

$$\begin{aligned} C_{O_2} &= 1.97 \times 10^{-7} P_{O_2} \exp\left(\frac{498}{T}\right) \\ C_{H_2} &= 9.174 \times 10^{-7} P_{H_2} \exp\left(\frac{-77}{T}\right) \end{aligned} \quad (9)$$

Notably,  $C_{O_2}$  is the oxygen concentration at the cathode/membrane interface and  $C_{H_2}$  is the hydrogen concentration at the anode/membrane interface.

- (iii) The ohmic overvoltage is given by,

$$V_{ohm} = IR_{int} \quad (10)$$

where the internal resistance  $R_{int}$ , obtained by the empirical analysis, is written as,

$$R_{int} = \frac{r_M I_m}{A_{fc}} \quad (11)$$

and the membrane resistivity  $r_M$  is given by

$$r_M = \frac{181.6 [1 + 0.03(I/A_{fc}) + 0.062(T/303)^2 (I/A_{fc})^{2.5}]}{[11.866 - 3(I/A_{fc})] \exp[4.18((T - 303)/T)]} \quad (12)$$

According to Eqs. (1)–(12), a single fuel cell is affected by current density, cell temperature, hydrogen and the partial pressure of oxygen. Table 1 presents parameter definitions and values of a Ballard 5 kW PEMFC stack system. Since all cells are in series, the total voltage for the stack is given by

$$V_{stack} = 35V_{fc} \quad (13)$$

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