Experimental Performance Evaluation of a Combustion Heat-Assisted Catalytic Flat Plate Fuel Reformer for Fuel Cell Grade Hydrogen

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ABSTRACT

Combining exothermic catalytic combustion heat on one side of a very high heat convective thin flat plate and endothermic catalytic reforming reaction on the other side, catalytic flat plate provides excellent heat integration into a compact fuel reformer system. Based on a two-dimensional computational fluid dynamics (CFD) model simulation results, a real world compact catalytic flat plate fuel reformer was built and experimentally evaluated its performance, for generating fuel cell grade hydrogen, in an actual high temperature polymer electrolyte membrane (PEM) fuel cell system. The two-dimensional CFD model helps to optimize various design parameters necessary to build the actual compact reformer and eliminated the uncertainties of heat and mass transfer coefficient values arise in one-dimensional CFD model. The performance of the compact catalytic flat plate fuel reformer was evaluated using a 5-cell high temperature PEM fuel cell stack at 160°C with reformat fed directly from the reformer to the anode stream of fuel cell stack. For performance comparison, a 5-cell high temperature PEM fuel cell stack was first evaluated with industry-standard pure hydrogen (0% CO) as well as with various percentage of CO mixed hydrogen (2%CO and 5%CO). Experimental results showed that if the average cell voltage drop within 0.03~0.05mV of a 5-cell high temperature PEM fuel cell stack would be acceptable then the reformat can be used at 160°C instead of pure hydrogen. The experimental results also indicated that the reformer will be able to provide actual fuel cell grade hydrogen that could be directly pumped from the compact catalytic flat plate fuel reformer into the high temperature PEM fuel cell stack.

KEY WORDS: Fuel Reforming, Fuel Cell Grade Hydrogen, Catalytic Flat Plate Reformer, Experimental Performance Evaluation, High Temperature PEM Fuel Cell.

Introduction

Significant amount of attentions are drawn recently on the rapidly growing fuel cell technology, such as proton exchange membrane fuel cell (PEMFC), because of its high potential to provide clean energy in various real-world industrial applications including transportation, auxiliary power supply and portable electronic devices [1-2]. PEMFC is considered as an electro-chemical device that converts chemical energy of hydrogen and oxygen directly into electrical energy without producing any greenhouse gases, since water and heat are the only byproduct produced through electrochemical reaction. Therefore, PEMFC is a pollution free clean energy source in which pure hydrogen is used as an anode reactant gas. PEMFC is also highly efficient due to only one step of energy conversion is needed compared to multiple step conversions are necessary for fuel combustion-based power generation technology, such as internal combustion (IC) engine, to produce useful electrical power [1-2]. But pure hydrogen as an anode reactant gas source for PEMFCs, at present, has a number of formidable limitations [1-2]. One of the major limitations is on-board hydrogen storage for transportation and mobile applications [2]. The onboard hydrogen storage issue is escalated by the lack of infrastructure for hydrogen distribution [2] system nationwide. Another fact is that there is no natural reserve for hydrogen in the earth. So, hydrogen must be extracted either from other natural resources through various means or could be produced on-board by reforming process [2-15]. As a viable alternative of carrying pure hydrogen and lack of hydrogen dispensing infrastructure, onboard hydrogen generation by reforming hydrocarbon fuels available in established nationwide refueling stations such as natural gas, gasoline, diesel, propane or alcohol-based bio-fuels would be an obvious choice [2-6, 8-17]. The only issue of using reformat fuel into the PEMFC stack is to degradation of PEMFC stack performance due to poisoning of Pt catalyst, used in PEMFC, by the carbon

monoxide (CO) presence in the reformat fuel [1-2]. Recently, development of temperature resistant high conductive polymer membranes, such as the acid-doped polybenzimidazole (PBI) membranes [2, 18-20], open up the opportunity to build high temperature PEM fuel cell (HTPEMFC) stack which can be fed with lower quality (i.e. higher CO concentrations in hydrogen) reformat hydrogen [2, 18-20] directly from the fuel reformer. From the thermodynamics point of view, the high operational temperature makes it possible to improve the CO tolerance in HTPEMFC stack at a higher level [2, 18-20]. Hydrogen-rich reformat can be produced on-board by reforming process using hydrocarbon fuels available at the nationwide refueling stations [5-6, 21-29]. However, the reformer should be highly efficient as well as compact in order to integrate the reformer on-board to feed the HTPEMFC systems.

Designing a compact fuel reforming system is remained always a major challenge, especially to ensure efficient distribution of heat on both the reforming and combustion sides during the fuel reforming process. The conventional tubular-type fuel reformer [21-22], as shown in Fig. 1, has long been used in the industry to reform hydrocarbon fuels with steam and catalytic reactors. As seen in Fig. 1a, the hydrocarbon gas mixture (fuel plus steam) has to be heated up using an external furnace and then distributed it into catalyst/reactor-filled reformer tubes. The gas mixture flows from top to bottom through tubes arranged in vertical rows. As the hydrocarbon fuel-steam mixture flew through the externally heated tubes - they react, produced hydrogen and carbon monoxide [21]. This system has lot of disadvantages such as higher heat loss due to lower heat transfer rate, higher cost and lower conversion efficiency [22]. In addition, as seen in Fig. 1b, there is a considerable amount of temperature drop, ΔT , between the furnace and the reactor side. Therefore, lot of heat energy is wasted while using this conventional type of reformer system [21-22]. To eliminate the excessive heat loss due to higher temperature drop in

conventional reformer, study on fuel reforming with catalytic combustion and steam reforming has also been studied [23-29].

Fig. 1 - Schematic of conventional fuel reforming process with tubular packed bed reactor [21-22].

As shown in Fig. 2, catalytic flat plate steam reformer is a compact alternative by combining the exothermic catalytic combustion reaction on one side of the thin plate coated with combustion catalyst and endothermic reforming reaction on the other side of the thin plate coated with reforming catalyst. The combustion reaction generates enough heat to drive the endothermic fuel reforming reaction on the reforming side of the plate through heat conduction [23-25]. In addition, catalytic fuel reformer plate can be arranged in stacks (see Fig. 2a) to build a suitable fuel reformer according to the need of specific applications. Many advantages can be achieved through catalytic flat plate fuel reformer design including compact standardized size (convenient

to use in automobile applications), lower cost, better conversion efficiency as well as higher heat transfer capacity [23-28] due to lower temperature drop, ΔT , as can be seen in Fig. 2b.

Fig. 2 - Schematic of catalytic flat plate fuel reforming process [23-26].

Moreover, fuel reforming efficiency increases in catalytic flat plate fuel reformer because of a very short distance heat exchanges between heat source (combustion side) and heat sink (reformer side). Additionally, lower operating temperature of the catalytic combustion helps low NO_x formation and reduces sudden temperature spikes and eliminates hot spot generations which are common in conventional fuel reformers [21-26]. Catalytic fuel reforming is a flameless process, so long radiation paths needed in conventional furnaces can be replaced by carving few millimeters diameter fuel channels on the plate geometry and hence it will dramatically reduce the reactor size [21-25]. Therefore, it is important to investigate the catalytic flat plate fuel reformer design in more details especially to study the heat distribution, heat transfer efficiency and ability to produce fuel cell grade hydrogen by the flat plate fuel reformer.

In this study, a catalytic flat plate fuel reformer was built based on the optimized two-dimensional computational fluid dynamics (CFD) simulation results [30]. The performance evaluation of the catalytic flat plate fuel reformer was carried-out using a 5-cell high temperature PEM fuel cell stack. Natural gas (i.e. Methane) was used as a fuel in the catalytic flat plate fuel reformer to generate hydrogen-rich reformat gas. Then reformat gas was directly pump into the 5-cell high temperature PEM fuel cell stack to evaluate the performance of the catalytic flat plate fuel reformer and assess the viability of producing fuel cell grade hydrogen generation. Details of the experimental measurements and investigations are presented in the subsequent sections.

Reaction mechanisms of natural gas reforming

Steam reforming of natural gas (i.e. methane) is a widely used process for industry-standard hydrogen gas production. The chemical reactions involved in reforming process are given below [30-31].

Initial steam reforming reaction:

Water gas shift reaction:

Steam reforming side reaction:

Both initial steam reforming reactions and the steam reforming side reactions are highly endothermic. Therefore, a catalytic exothermic combustion reaction is needed on the other side

of the flat plate wall to provide the necessary heat to drive the reforming reactions in the reforming side of the plate. Combustion side reaction of methane is given below.

Combustion reaction of methane [30-31]:

The reaction rate equations for the chemical reactions in reforming and combustion sides are considered as follows [23, 30].

Initial steam reforming reaction rate:

$$r_1 = \frac{\frac{k_1}{p_{H_2}^{2.5}} * \left[p_{CH_4} * p_{H_2O} - \left(\frac{p_{H_2}^3 * p_{CO}}{K_{e,1}} \right) \right]}{(Den)^2} (mol / kg_{cat} / s) \quad (5)$$

Water gas shift reaction rate:

$$r_2 = \frac{\frac{k_2}{p_{H_2}} * \left[p_{CO} * p_{H_2O} - \left(\frac{p_{H_2} * p_{CO_2}}{K_{e,2}} \right) \right]}{(Den)^2} (mol / kg_{cat} / s) \quad (6)$$

Steam reforming side reaction rate:

$$r_3 = \frac{\frac{k_3}{p_{H_2}^{3.5}} * \left[p_{CH_4} * p_{H_2O} - \left(\frac{p_{H_2}^4 * p_{CO_2}}{K_{e,3}} \right) \right]}{(Den)^2} (mol / kg_{cat} / s) \quad (7)$$

Methane combustion reaction rate:

$$r_4 = k_4 * \frac{p_{CH_4}}{R * T} * \exp\left(\frac{-90000}{R * T}\right) (mol / m^3) \quad (8)$$

where

$$DEN = 1 + K_{CO} * p_{CO} + K_{H_2} * p_{H_2} + K_{CH_4} * p_{CH_4} + \frac{K_{H_2O} * p_{H_2O}}{p_{H_2}} \quad (9)$$

All the values of pre-exponential factors (k_i , $i=1,...,4$, CO, H₂O, CH₄, H₂), heat of adsorption, activation energies and equilibrium constants ($k_{e,i}$; $i=1,2,3$) are given in [23, 30].

Computational modeling and simulation procedure

A two dimensional computational fluid dynamics (CFD) model [30] containing three sub domains such as reformer section, flat plate, and combustion section as shown in Fig. 3 is simulated with 0.3m length of each sub-domain. Reforming sub-domain section is extended to additional 0.3m in length in order to complete the water gas shift reaction after initial fuel reforming. Flat plate thickness and reforming as well as combustion channel heights were considered as 5mm and 1.1 mm respectively.

Fig. 3 - Schematic of computer model domain geometry [30].

Steady-state 2D incompressible Navier-Stokes equation, Stefan-Maxwell multi-component diffusion and general heat transfer equation are used for momentum transport, species transport and energy transport for both reformer and combustion sections. Furthermore, we assumed all the reactant gases follow ideal gas law, $p = \rho RT$. Model governing equations for gas phase and flate plate can be written as [30-31]:

$$\text{Continuity: } \nabla(\rho u) = 0 \quad (10)$$

$$\text{Momentum transport: } \rho(u \cdot \nabla u) = -\nabla p + \mu \nabla^2 u \quad (11)$$

$$\text{Species transport: } \nabla \left\{ -\rho^* w^n * \sum_k D_k^n * \left(\nabla x_k + (x_k - w_k) \frac{\nabla p}{p} \right) \right\} = R - \rho^* u^* \nabla w^n \quad (12)$$

$$\text{Heat transfer for reforming and combustion: } \nabla (-k^* \nabla T) = Q - p C_p^* u^* \nabla T \quad (13)$$

$$\text{Heat transfer (flat plate): } \nabla (-k^* \nabla T) = 0 \quad (14)$$

where $n \neq k$, n and k indicate different species, R is the reaction rate, u is the velocity, w is the mass fraction of species, ρ is the density, p is the pressure, Q is the heat source, T is the temperature, k is the thermal conductivity, C_p is the specific heat at constant pressure and x is the species composition. To solve equations (10)-(14), the required boundary conditions are defined as [30]:

$$\left. \begin{array}{l} u = 0 \text{ (no slip condition at the wall)} \\ \text{Inlet: } u = T = w = p = \text{prescribed condition} \\ \text{Outlet: } \frac{\partial w}{\partial y} = \frac{\partial T}{\partial y} = \frac{\partial u}{\partial y} = 0 \text{ (zero flux at the outlet)} \\ p = \text{atmospheric pressure condition} \end{array} \right\} \quad (15)$$

The diffusion coefficients for the multi component mixtures in reformer and combustion section were calculated using the Lenard Jones force constants [23, 30-31] defined as

$$(D_A^B)_0 = \frac{(0.001858) * T_0^{3/2} * \left[\frac{1}{M_A} + \frac{1}{M_B} \right]^{\frac{1}{2}}}{P * \sigma_{AB}^2 * \Omega_D} \quad (16)$$

$$\sigma_{AB} = \left(\frac{\sigma_A + \sigma_B}{2} \right) \quad (17)$$

$$\Omega_D = -0.0946459 * \left(\frac{\epsilon}{kT_0} \right)^2 + 0.909766 * \left(\frac{\epsilon}{kT_0} \right) + 0.637098 \quad (18)$$

$$\left(\frac{\epsilon}{k} \right)_{AB} = \left[\left(\frac{\epsilon}{k} \right)_A * \left(\frac{\epsilon}{k} \right)_B \right]^{0.5} \quad (19)$$

$$D_A^B = (D_A^B)_0 * \left(\frac{T}{T_0} \right)^{1.75} \quad (20)$$

where $T_0 = 273^\circ\text{K}$ is the absolute temperature. The molecular weights of components A and B are M_A and M_B . P is in atmospheric pressure. σ_A , σ_B and ε/k are the constants [31]. T denotes the temperature in the reformer or combustion section. Diffusion coefficients for the catalyst layer were calculated using the following equations.

$$D_{eff} = \frac{\varepsilon}{\tau} \left(\frac{1}{D_k} + \frac{1}{D_G} \right)^{-1} \quad (21)$$

$$D_k = 97 * R_p \sqrt{\frac{T}{M}} \quad (22)$$

D_k is the Knudsen diffusion coefficient, R_p is the pore radius, ε is the porosity of catalyst layer and τ is the tortuosity factor.

$$D_G = (D_A^B)_0 * \left(\frac{T}{T_0} \right)^{1.75} \quad (23)$$

In order to compare the simulation results with the conventional reformer [21-25], a porosity of 0.4 with pore radius of 10nm and tortuosity factor of 0.4 were used in the simulation for both combustion and reforming catalysts. In the simulation, steady state fully developed laminar flow profile was employed [30] at the inlet of both endothermic and exothermic channel, and reaction rates were considered as volumetric source terms instead of boundary conditions in both reformer and combustion sections. We assumed that both reformer and combustion domains are symmetric along the centerline of the domain. We used very thin (20-70microns) layer of reforming and combustion catalyst, and hence it is assumed to be isothermal in the transverse direction. The CFD model equations were solved using the appropriate boundary conditions described above and obtained optimized design parameters of catalytic flat plate fuel reformer.

Detailed results of design parameters study using the CFD model can be found in our earlier publication [30].

Experimental performance evaluation of the reformer using high temperature PEM fuel cell stack

Experimental set up of catalytic flat plate reformer

Based on the CFD model simulation results, a catalytic flat plate fuel reformer was built using the optimized design parameters. In order to evaluate the performance, the catalytic flat plate fuel reformer was hooked up with a 5-cell high temperature PEM fuel cell stack. To generate reformat, the inlet conditions for both reformer and combustion side of the reformer were set similar to those used by [23, 30]. However, atmospheric pressure condition was applied at the reformer outlet instead of operating at high pressure condition (29 bar) used in [23]. For water gas shift reforming, steam to gas (methane) ratio of 3.5 was used that corresponds to the typical industrial reforming condition [21-22]. Initially, fuel (methane) and air were preheated to 793°K and supplied to the reformer (fuel only) and combustion section (fuel and air) simultaneously. It can be noted that the initial temperature profile is comparable to the temperature profile of a conventional reformer [22]. The most common industrial catalyst used for the reactions on reformer side is based on Ni on alumina support. The reforming catalyst that contains 15.2% Ni supported on magnesium spinel was used which was similar to the catalyst used by [23, 30]. The same amount of catalyst loading of 2355kg/m³ was used both in the simulation [30] study and in the present experiment with catalytic reformer. The activation energy for methane catalytic combustion varies from 75 to 200kJ/mol. The most common catalysts used for catalytic combustion of methane are supported precious metal catalysts such as Pt, Pd, and Rh. The

activation energy depends on the catalyst used for the combustion reaction. Following the results of a simulation done by [23, 30], the activation energy of 90kJ/mol was used both in the simulation and experiment. It was assumed that 99.99%~100% methane conversion were taken place in the reforming section and also the amount of methane delivered into the combustion chamber were burnt completely (i.e. 100%) through combustion. Through exothermic reaction, combustion chamber produced heat that eventually transferred to the reforming side by the flat plate conduction and complete the endothermic reaction processes on the reforming side to produce reformat gas. Finally, through the water gas shift reaction section, the reformat gas got further purified and resulted in a very low amount of CO in the reformat gas.

Experimental set up of 5-cell high temperature PEM fuel cell stack and testing procedure

A 5-cell high temperature PEM fuel cell (HTPEMFC) stack with phosphoric acid-doped PBI membrane was attached with the catalytic flat plate fuel reformer. Once the attachment/installation was completed, the 5-cell HTPEMFC stack was tested in this investigation with three different anode fuels along with the reformat gas obtained from the catalytic flat plate fuel reformer and study the current-voltage characteristics of the HTPEMFC. Besides the reformat gas, the three other fuel grades were: i) industrial grade pure hydrogen (0% CO), ii) a certified gas mixture consisting of 1.99% CO (i.e. 2%CO) balanced hydrogen, and iii) a certified gas mixture consisting of 4.99% CO (i.e. 5%CO) balanced hydrogen. All three fuels were supplied from compressed gas cylinders and delivered to the 5-cell stack using a MKS mass flow controller. Gas mixtures were certified by the gas supplier using a gravimetric method. The reformat gas was directly pumped from the catalytic flat plate fuel reformer to the HTPEMFC. We set, 1.2 fuel stoichiometry with 0.1 *slm* minimum fuel flow rate at the anode

inlet and 2.5 air stoichiometry with 0.3 *slm* minimal air flow rate at the cathode inlet and atmospheric (ambient) pressure was maintained at the cell outlet (exit). First the HTPEMFC stack temperature was raised from room temperature to 130°C and supplied industrial grade pure hydrogen (0% CO) to the anode and then allowed the stack to reach equilibrium. For measuring cell temperature we used thermocouple sensors. The actual measured temperature was within 130°C ~132°C. When the stack voltage had reached steady state at 130°C, polarization curves were measured. It should be noted that at the time the polarization curves were measured, the HTPEMFC stack had already operated for over 1200 hours. Once the polarization curve measurement was finished, the stack was set to operate at constant current in order to avoid OCV (open circuit voltage). The above-mentioned process was repeated for 140°C, 150°C and 160°C, and measured the polarization curves at steady state for each of the cases. It was observed that the actual stack temperature was within +2°C at the time of polarization curve measurement. The above-mentioned procedures were repeated for 2% CO, 5% CO and reformat gas mixtures, and measured the HTPEMFC stack polarization curves for each gas mixture.

Results and discussions

Evaluation of optimized reformer design based on CFD modeling results

The experimental performance evaluation of the newly built catalytic flat plate reformer, based on optimized design parameters obtained from our 2D computational fluid dynamics (CFD) model simulation results, are presented and discussed in this section. Since catalytic flat plate fuel reformer use combustion heat from one side of the flat plate to reform the fuel in the reforming section of the reformer, it is important to know the detailed temperature distribution across the catalytic flat plate fuel reformer.

Fig. 4 - Temperature distribution along the channel length across all three regimes – combustion, flat plate and reforming sections. Blue color represents applied inlet temperature of 790°K and red color represents steady state temperature of 1115°K at the channel end.

Fig. 4 represents the temperature distribution along the channel length across all three sections - combustion, flat plate and reforming section of the reformer. Blue color represents the applied inlet temperature and red color represents the maximum temperature after combustion heat transported into the reforming section through the flat plate heat conduction. From Fig. 4, it can be seen that as the combustion heat produce along the channel it quickly transported into the reforming side of the reformer to complete the endothermic fuel reforming reactions (see equations (1) and (3)). Temperature distribution reached at a steady state once the fuel reforming reaction had completed at the channel end. From Fig. 4 we also see that the temperature of the combustion heat reached at 1115°F at the channel end compared to the applied channel inlet temperature of 790°F. As seen in Fig. 4, a little bit temperature variations were observed between

the combustion and reforming sides of the catalytic flat plate fuel reformer especially within the middle zone of the channel length, i.e. between the channel length of 0.05m~0.2m. In order to quantify the exact temperature variation between combustion and reforming sides of the catalytic flat plate fuel reformer, we computed transverse temperature profile from the temperature distribution shown in Fig. 4.

To assess the effectiveness of heat transfer process across the flat plate by heat conduction, it is very important to examine transverse temperature profile for both reformer and combustion section. The transverse temperature profile was measured as the temperature difference of the gas between the flat plate wall and the channel center of fuel reformer (or combustion) section, $\Delta T_g = T_{\text{wall}} - T_{\text{center}}$. Fig. 5 represents the measured transverse temperature profiles for both combustion and reformer channels. It can be clearly seen from Fig. 5 that the transverse temperature profile is almost the same upto 0.06m, which is 20% of the reformer/combustion channel length, for both sides of the flat plate. It is due to the fact that the flat plate conduction is occurring in these regions of the channel with virtually negligible thermal resistances in order to transfer heat produced by the combustion reaction quickly to the reformer side. From Fig. 5, it can be observed that the maximum temperature difference was about 61°K at the reformer side whereas it was about 78°K at the combustion side. A noticeable temperature gradient was observed between the channel length of 0.06m and 0.2m. It was due to the change of thermal resistance of the plate material during the increased heat generation by the high intensity of combustion reactions in the combustion side. Temperature in both the combustion and reformer side had reached steady state due to saturation of both reforming and combustion reactions, as per equations (1)~(4), at the tailing end of the channel.

Fig. 5 - Measured transverse temperature profile in the gas phase of combustion and reformer sides, $\Delta T_g = T_{\text{wall}} - T_{\text{center}}$.

In order to build a real-world catalytic flat plate fuel reformer, we used optimized reformer design parameters obtained from a 2D CFD model simulation results with water gas shift (WGS) reaction by taking into account the effect of channel length, inlet gas velocity and catalyst layer thickness of the flat plate reformer system [30]. The CFD model equations and simulation method are discussed in the section 3 above and also can be found in our earlier publication [30]. While using optimized reformer geometry, at the end of initial reforming process (i.e. before WGS reaction start) we observed that the reformat gas temperature was at around 1115°K. To achieve maximum conversion efficiency with WGS reaction catalyst,

reformat gases need to have temperature at around 800°K at the end of initial fuel reforming process. In order to comply with these requirements, we used a heat exchanger to bring down the reformat gas temperature from 1115°K to 800°K before it entering into the water gas shift reaction zone of the reformer. It can be noted that the excessive heat extracted through heat exchanger can be utilized to heat up the reformer inlet gas/fuel (for preheat) which will eventually improve the overall system efficiency. We observed that no methane was present after the WGS reaction was completed (i.e. 100% methane was converted during WGS reforming reaction). In the WGS reaction zone, carbon monoxide present in the reformat gas after initial reforming reacts with excess steam and left-over methane in the reformat gas to produce carbon dioxide (CO₂) and hydrogen as per equations (1) ~ (3). The WGS reaction is mildly exothermic. Therefore, it is very important to maintain WGS reaction zone temperature at 800°K for efficient WGS reaction activity. Fig. 6 represents the production of dry H₂ after completion of WGS reaction with optimized reformer geometry. From Fig. 6 we can see that the production of dry hydrogen went up slightly from 0.76 mole after initial reforming to 0.78 mole after completion of WGS reaction i.e. by only 2% with the WGS reaction. It may due to the fact that very little amount of methane was present in the reformat gas after initial reforming and hence a very little amount of methane was available in the WGS zone to produce H₂. Fig. 7 represents production of dry CO during the WGS reaction with optimized reformer geometry obtained from model simulation results. The amount of dry CO went down more than 90% with WGS reaction as can be seen from Fig. 7. Therefore, it was justified to include WGS reaction with the

Fig. 6 - Production of dry H_2 during WGS with optimized reformer geometry [30].

Fig. 7 - Reduction of dry CO during WGS with optimized reformer geometry [30].

optimized fuel reformer geometry in order to get reduced or low CO level reformat H₂. It is important to note that PEM fuel cell performance is very susceptible to CO present in the anode fuel stream because of Pt catalyst poisoning. It is due to the fact that platinum (Pt) catalyst is used in the PEM fuel cell for electro-kinetic reactions to produce useful electric power. CO poison Pt catalyst as per the thermo-absorption and chemisorption properties of Pt [18]. So, it is necessary to make sure that the reformat gas contains very low level of CO in order to avoid Pt poisoning during HTPEMFC stack operation with reformat gas.

Experimental performance evaluation of the reformer using 5-cell high temperature PEM fuel cell stack

Fig. 8 represents the measured polarization curves at different temperatures of a 5-cell high temperature PEM fuel cell stack operated with 0~5%CO mixed hydrogen as well as with reformat gas fed directly from the catalytic flat plate reformer to the anode stream of fuel cell stack. From Fig. 8, we can see that for the case of reformat, there was virtually no fuel cell stack voltage drop up to current density of 0.25A/cm² as compared to pure hydrogen fed at 160°C. When the stack was fed with pure hydrogen and reformat at 160°C the voltage drop was within 0.03~0.05mV at high current density of 0.4A/cm² as can be seen from Fig. 8. It is also found from Fig. 8 that the HTPEMFC stack voltage drop was much lower with reformat fed at 160°C as compared to 2%CO and 5%CO fed at the anode fuel stream especially at high current density of 0.3~0.4A/cm². This result clearly indicates that the direct feed of reformat gas from the reformer would be more preferable at 160°C than 2%CO~5%CO fed in the HTPEMFC stack operation.

Fig. 8 - Average cell voltage of 5-cell stack high temperature PEM fuel cell as a function of cell current density measured at different temperatures with different CO concentrations mixed hydrogen as well as with the reformat directly fed from the reformer.

In order to quantify amount of voltage drop of HTPMFC stack for using reformat gas as well as 2%CO and 5%CO mixed hydrogen instead of pure hydrogen, we plotted the average cell voltage drop as a function of current density. The HTPMFC average cell voltage drop is calculated by the average cell voltage obtained using industry-standard pure hydrogen (i.e. 0%CO mixed hydrogen) minus the average cell voltage obtained using reformat gas as well as 2%CO and 5%CO mixed hydrogen, respectively. Fig. 9 represents the measured average cell voltage drop as a function of current density at 160°C using reformates and 2% ~ 5%CO mixed hydrogen. From Fig. 9, we can see that no cell voltage drop observed for reformer feed until current density of 97mA/cm² whereas cell voltage drop of 0.005~0.02mV was observed for

2%CO and 5%CO mixed hydrogen. As stack current density increased the average cell voltage drop also increased.

Fig. 9 - Measured average cell voltage drop in a 5-cell stack high temperature PEM fuel cell stack fed with reformat gas as well as 2%CO and 5%CO mixed hydrogen as compared to industry-standard pure hydrogen (0%CO) fed at 160°C.

As shown in Fig. 9, the average cell voltage loss was very high for 5% CO compared to 2% CO and reformat gas at 160°C at higher current densities, 300~450 mA/cm^2 . Although the cell voltage loss was relatively small for 5% CO and 2% CO up to current density 98 mA/cm^2 at 160°C, the cell voltage loss was increased after current density 100 mA/cm^2 for 5% CO and 2% CO compared to reformat gas at 160°C. If we consider the average cell voltage loss within less than 0.05mV is acceptable, then 2% CO and reformat gas can be used at 160°C as can be seen

from Fig. 9. From Fig. 9, it can be clearly seen that the average cell voltage loss was the lowest for reformat gas feed compared to 2%CO and 5%CO reactant gas feed. It indicates that reformat can be directly fed in the HTPEMFC stack instead of pure hydrogen.

Fig. 10 - Measured average cell power loss, ΔW (mW), obtained experimentally in a 5-cell stack high temperature PEM fuel cell stack fed with reformat gas as well as 2%CO and 5%CO mixed hydrogen as compared to industry-standard pure hydrogen (0%CO) fed at 160°C.

Fig. 10 presents the average cell power loss as a function of current density for the reformat as well as for 2% CO and 5% CO in the hydrogen stream at 160°C temperature. The power loss is calculated by the difference between cell power obtained using industry-standard pure hydrogen (0%CO) and reformat or CO-containing hydrogen (i.e. 2% ~ 5%CO mixed

hydrogen) at different current densities. From Fig. 10, there were no cell power losses with reformat within 96mA/cm^2 whereas we see that 0.005mW power losses occur for both 2% CO and 5% CO at 160°C at current densities within 96mA/cm^2 . At 160°C , power losses are higher for 5% CO as compared to 2% CO and reformat gas feed at current density above 100mA/cm^2 . At 160°C , power losses are the highest for 5% CO as compared to 2% CO and reformat gas feed at higher current densities, $150\sim 215\text{mA/cm}^2$. From the electrochemistry analysis discussed in Das et al [18] and the results presented in Das et al [18], it was revealed that the cell voltages as well as power losses were apparently a function of the CO content in the anode hydrogen stream, temperature, anode catalyst and current density. The results presented in Figs. 9 and 10 agreed quite well with the observation and results reported in Das et al [18]. From Fig. 10, it was observed that the reformat gas feed can be tolerated at 160°C when the average cell power loss/drop within 0.025mW would be acceptable at higher current densities. From Fig. 10, it can also be seen that the average cell power losses were the lowest for the reformat gas fed directly from the catalytic flat plate reformer compared to 2% and 5% CO mixed hydrogen fed in the anode. It implies that the reformat can be used in HTPEMFC stack instead of pure hydrogen in the anode at 160°C .

Conclusions

Based on the results of an optimized CFD model geometry simulation with water gas shift reactions and optimized design parameters those control the performance and efficiency of a fuel reformer, a combustion heat assisted catalytic flat plate fuel reformer was built and experimentally evaluated its performance in this investigation. Water gas shift reactors were used and incorporated in the catalytic flat plate fuel reformer in order to increase hydrogen-rich

reformat production and to reduce the CO level significantly in the reformat gas as can be seen from Figs. 6 and 7. Combustion heat transfer through the thin flat plate into the reforming side was almost the same at the beginning and end of the fuel channel with a very small temperature gradient, at about 71°C, in the middle section of the channel as shown in Figs. 4 and 5. It implied that the heat transfer from the combustion section into the reformer section of the catalytic flat plate fuel reformer was highly efficient. The performance of the catalytic flat plate fuel reformer was evaluated by directly supplied the reformat gas from the reformer into a 5-cell high temperature PEM fuel cell stack and measured the cell as well as stack voltages with applied current loads. For comparison, industry-standard pure hydrogen (i.e. 0%CO), 2%CO and 5%CO mixed hydrogen also fed into the 5-cell HTPEMFC stack and measured the voltages with applied current loads. Experimentally measured results showed that the average cell voltages with reformat gas were almost the same as of pure hydrogen feed for the entire range of cell current densities at 160°C as can be seen from Fig. 8. The current-voltage curves (i.e. polarization curve) of the 5-cell stack shows upward trend with reformat feed compared to 2%CO and 5%CO mixed hydrogen feed. It implies that the direct reformat feed from the reformer performed much better than 2%CO and 5%CO mixed hydrogen feed at the anode. The diagnostic evaluation of 5-cell HTPEMFC stack showed that the reformat can be used at 160°C if the average cell voltage drop within 0.03~0.05mV would be acceptable at high current densities compared to 0.04~0.11mV voltage drop for 2%CO and 5%CO mixed hydrogen as per Fig. 9. The experimental measurement of average cell power losses of the 5-cell HTPEMFC stack also revealed that the reformat can be used instead of pure hydrogen at 160°C if the average cell power loss of 0.01~ 0.025mW would be acceptable at higher current densities compared to average cell power losses of 0.02~ 0.045mW for 2%CO and 5%CO mixed hydrogen. Finally,

based on the experimental measurement results using a 5-cell HTPEMFC stack with pure hydrogen, reformat gas feed directly from the reformer, 2%CO and 5%CO mixed hydrogen feed, it was clearly shown that, instead of using pure hydrogen, H₂-rich (low CO content) reformat gas can be directly fed from the optimized catalytic flat plate fuel reformer into the high temperature PEM fuel cell stack without further removal of CO from the reformat. Furthermore, if we considered the fact that a catalytic flat plate fuel reformer is a consumer of heat and water, and high temperature PEM fuel cell stacks are producer of heat and water, then the integration of HTPEMFC stack with the reformer should expected to improve the overall system efficiency and performance.

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