

Critical flow rate of anode fuel exhaust in a PEM fuel cell system[☆]

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Abstract

A manual purge line was added into the exterior fuel exhaust stream of a Ballard PEM stack in a Nexa™ power module. With the addition of manual exhaust purge, high levels of inert gases were intentionally added to the anode feed without changing normal operational procedures. A new method of determining the critical minimum flow rate in the anode exhaust stream was given by an anode mass balance. This type of operation makes dual use of membranes in the MEAs as both gas purifiers and as solid electrolytes. The PEM stack was successfully operated with up to ca. 7% nitrogen or carbon dioxide in the absence of a palladium-based hydrogen separator at ca. 200 W power level. Nitrogen in the anode stream was concentrated from 7.5% to 91.6%. The system maintained a fuel efficiency of 99% at a manual purge rate of 2.22 ml s⁻¹ and no auto purge. The fuel cell stack efficiency was 64% and the stack output efficiency was 75%. The overall system efficiency was 39%. After troublesome CO and H₂S poisons were removed, a hydrocarbon reformat containing high levels of CO₂ and H₂O was further used in the Nexa™ stack. The size and complexity of the fuel processing system may be reduced at a specified power level by using this operational method.

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1. Introduction

The PEM fuel cell system is an emerging power source providing high power density, high energy conversion efficiency, and cleaner emissions than many other available technologies [1]. The system has many applications such as generating electricity, supplying hot water for baths, and heating cold room air by recovering heat from the electrochemical reactions. Its fuel gas at the anode side undergoes an electrochemical oxidation of hydrogen by losing electrons, while the exhausted inert gas and water vapor (or liquid) accumulate at the exit portion of flow channel. Oxygen is electrochemically reduced with hydrated protons at the cathode side, producing water and heat. Further co-generation or fuel recycle on the system operation of the polymer electrolyte fuel cell (PEFC)

can be helpful for obtaining higher fuel and overall energy efficiency [2].

For a complete fuel cell system, the management of small amounts of exhaust gas in the anode side is significantly related to the system's energy efficiency and its operation lifetime. Most work on mathematical models and structure optimizations have been based on a unit PEM fuel cell [3]. No model and related work was found in the literature that addresses the anode gas flow and its exhaust in a stack and integrated system. A fuel cell stack, including its control system, needs various optimal methods to operate and convert chemical energy to electricity as efficiently as possible for different fuel sources [4,5]. Wells et al. [6] gave a resuscitation method and apparatus for which there was a decrease in stack performance. The system and its stack kept running rather than performing an emergency shutdown, if the fuel cell system successfully carried out this resuscitation process. Barton et al. [7] developed an apparatus for stack exhaust purge using purge conditions determined by comparing the performance of one or more fuel cells in a purge

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Nomenclature

E	voltage (V)
f	frequency (Hz)
F	Faraday's constant, $96487 \text{ As mol}^{-1}$
ΔG	Gibbs free energy for electricity generation (kJ mol^{-1})
ΔH	thermal energy (enthalpy change) available from the H_2/O_2 reaction (kJ mol^{-1})
I	current through fuel cell stack (A)
m	total number of the working fuel cells in the stack
n	number of electrons involved in one water molecule production
q	heat produced in the reaction which is contributed to useful work for energy saving (kJ mol^{-1})
u	gas flow rate at the inlet side in equations (mol s^{-1} , i.e. $2.404 \times 10^4 \text{ ml s}^{-1}$ at 20°C , 1 atm)
v	gas flow rate at the outlet side in equations (mol s^{-1} , i.e. $2.404 \times 10^4 \text{ ml s}^{-1}$ at 20°C , 1 atm)
x	outlet exhaust volume fraction
X	fuel inlet volume fraction

Greek letter

η	efficiency
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Subscripts

0	fuel gas inlet
1	hydrogen fuel gas
2	inert gas
a	auto purge outlet
e	main channel exit of the fuel cell anode
fc	fuel cell
fe	fuel cell energy
fu	fuel gas
h	manual exhaust outlet (hand adjustment)
j	number of the fuel cell started from the fuel inlet
oe	overall efficiency of the fuel cell system
p	auto purge
s	fuel cell stack
so	stack output
w	useful work

cell portion of the stack with one or more cells in the stack. As a whole, gas (fuel, oxidant, and exhaust) management, water (liquid and vapor) management, thermal (heat transfer) management, and power (energy) management are the four main areas for PEFC system design and safe operation. Water and thermal management exhibit a close relationship

that has been widely investigated in order to develop MEAs and operate the PEFC system safely [8,9]. Gas management is the primary design issue of the fuel cell system for both MEA membranes and system safety concerns. This is essential for the system to maintain operation after start-up and to prolong its operational lifetime. In this paper, the fuel gas in the anode side and its exhaust management are discussed in detail and the critical flow rate of anode exhaust is determined by suitable manual adjustment of the exhaust flow rate.

2. Experimental

A PEM fuel cell stack in a NexaTM power module (Ballard Power Systems, SN00751) was tested in the laboratory. The NexaTM power module is an automated PEFC system, providing unregulated dc power through the use of an external hydrogen fuel supply. Its operation is limited only by the fuel purity requiring no more than 0.01% of total inert gases at a rated power of 1200 W. Hydrogen and air are supplied to two separate gas channels flowing through the flow field plates separately. The above PEM stack has a total of 47 MEAs (or cells) connected in series through 48 flow-field plates. The PEM stack and experimental schematic drawing are shown in Fig. 1. The operating pressure at the fuel supply inlet was chosen to be 20–40 psig. The fuel supply pressure to the stack was 5.0 psig, and the pressure of air oxidant was defined as 2.2 psig. The power system was air cooled and used no outside fuel or oxidant humidification. The lab test was conducted in a stack life time of no more than 78 h (the product operating life time is no less than 1500 h).

This system has its advantages: requiring low maintenance and being fully automated, portable, and highly integrated. The disadvantage of the system is that utilization of hydrogen feeds with high levels of inert gases or reformed gases is not convenient, because frequent purging for system restoration wastes fuel energy resulting in efficiency loss. It may cause system failure when the exhaust is released too frequently. Therefore, a manual purge line was added into the end of the NexaTM fuel exhaust line in order to use the system for the anode fuel and exhaust experiment. Making use of the auto purge line, the fuel cell system was able to be normally started and shut down safely. By using a back pressure valve, the manual purge stream was separated from the NexaTM exhaust line. A gas bubble meter was connected to the manual exhaust stream for accurate measurement of the gas flow rate (Fig. 2). The PEM fuel cell system was first fed with pure hydrogen fuel (>99.99%) according to the product operation manual. After the fuel cell system was started at ca. 200 W stack load and allowed to reach a steady state at a constant current after approximately 20 min, the hydrogen fuel with inert gas was fed into the fuel cell system replacing the pure hydrogen fuel. The purge cell voltage was recorded by the NexaMon software (OEM 2.0 version) from Ballard. The Agilent 6890 gas chromatography system with enhanced integrator was utilized to measure the compositions of the fresh and exhaust

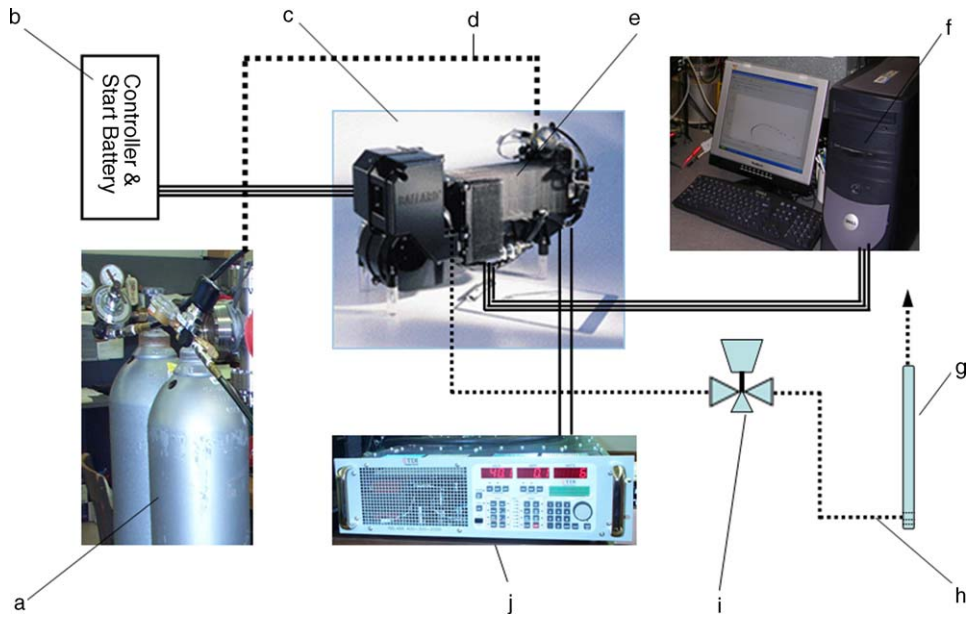


Fig. 1. Schematic drawing of a Nexa™ PEFC stack added with a manual purge line: (a) fuel gas cylinders, (b) central controller unit and start battery, (c) Nexa™ power system, (d) fuel gas line, (e) Ballard® stack, (f) computer data collection, (g) bubble meter, (h) gas exhaust line, (i) back pressure valve, and (j) electronic load.

gas streams. The flow rate was determined for feeds of either pure hydrogen or high levels of inert gases.

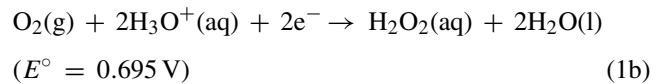
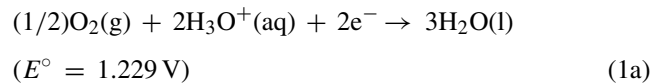
Higher polarization resistance exists at the cathode side rather than the anode side. In addition, water is formed at the cathode side, because catalyst-activated oxygen atoms react with protons to generate heat and water by taking electrons originally generated at the anode. This means that heat and water are mainly produced at the cathode side; thus, the effect of water vapor in the gas stream at the anode side is neglected when the fuel cell system is operated at 200 W stack power and ca. 30 °C stack temperature. Electronic loads (RBL488

Dynaload® Division and 6060B HP Electronic load) were applied to maintain a steady stack current.

3. Results and discussion

3.1. Fuel gas and its exhaust management in the anode stream

Gas (hydrogen fuel, air/O₂ oxidant, and their exhausts) management is an important issue for the design and operation of a complete PEM fuel cell system. Management of the fuel gas distribution and its exhaust stream(s) plays an important role in the operation of the PEM stack. This process is also related to the fuel efficiency and the overall system efficiency. The reaction in the aqueous acid at the cathode is the reduction of oxygen in air by receiving electrons as follows:



Some water produced at the cathode undergoes mass transfer by back-diffusion to the anode side for hydrogen electro-oxidation. The reaction at the anode is the oxidation of hydrogen by releasing electrons to the conductive electrode and on through the external load:

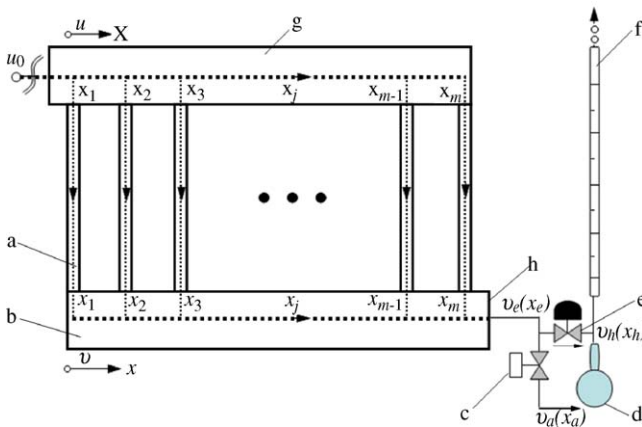
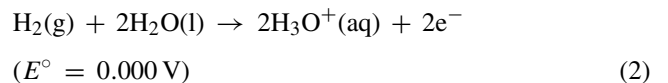


Fig. 2. Simple flow diagram of anode fuel and exhaust stream in a PEFC stack: (a) fuel cell anode and fuel flow, (b) main exhaust flow channel, (c) auto purge valve (average flow rate expressed as v_a), (d) bubble generator, (e) back pressure valve, (f) bubble meter (flow rate expressed as v_h), (g) main fuel gas channel (feed for each cell assumed in parallel), (h) exhaust outlet (flow rate expressed as v_e).

The protons produced exist as hydronium ions, which move toward the highly proton conductive polymer membrane. Most protons diffuse through the hydrated membrane, while the rest of hydrated protons are transported to the cathode by electroosmotic drag [1]. Water vapor in the gas exhaust streams, especially at the anode side, is neglected due to the low stack operating temperature (ca. 30 °C). The inlet flow rate can be directly measured by a mass flow meter/controller, or calculated using the measured exhaust flow rate and compositions of the inlet and the exhaust gases.

For a simple exhaust analysis, the PEM fuel cell system was operated at a constant current ranging from 50 to 200 W of stack power. After the fuel gas stream flows into the common fuel cell inlet, it is assumed to be uniformly distributed to every flow field plate and anode unit, i.e., $X_{01} = X_{j1}$ ($j = 1-47$) for hydrogen fuel and $X_{02} = X_{j2}$ ($j = 1-47$) for inert gas. The hydrogen fuel is approximately consumed at an equal rate in each cell. At each cell outlet, the gas composition is assumed equal. The exhaust line in the main flow channels is shown in Fig. 2. The pressure drop is quite small in both main flow channels. The small amount of gas crossover from each electrode is neglected in this work. It is further assumed that the gas composition is the same at the auto purge and the manual purge outlets. Also, the auto purge frequency (f_p) is assigned a value of ∞ , which means the exhaust gas is purged equally at a small average flow rate. Thus, the total continuous volume is considered the same as that of exhaust gas purged during normal operation ($0 < f_p < \infty$). From a mass balance and the above mentioned assumptions, the following equations can be constructed:

$$x_{a1} = x_{h1} \quad (3)$$

$$x_{a2} = x_{h2} \quad (4)$$

$$x_{e1} = x_{h1} \quad (5)$$

$$x_{e2} = x_{h2} \quad (6)$$

$$X_{01} + X_{02} = 1 \quad (7)$$

$$x_{h1} + x_{h2} = 1 \quad (8)$$

$$v_a + v_h = v_e \quad (9)$$

$$u_0 X_{01} - v_e x_{e1} = \frac{mI}{2F} \quad (10)$$

$$u_0 X_{02} = v_a x_{a2} + v_h x_{h2} \quad (11)$$

Combining Eqs. (3)–(11), the inlet/outlet flow rates and the average flow rate of auto purge are then obtained:

$$u_0 = \frac{x_{h2} m I}{(X_{01} - x_{h1}) 2 F} \quad (12)$$

$$v_e = \frac{1 - X_{01}}{X_{01} - x_{h1}} \frac{m I}{2 F} \quad (13)$$

$$v_a = \frac{1 - X_{01}}{X_{01} - x_{h1}} \frac{m I}{2 F} - v_h \quad (14)$$

Flow rates shown in the above equations are in mol s^{-1} . The rates are converted to ml s^{-1} by using the ideal gas assumption at 1 atm and 20 °C for convenient description in the text and the table. The average flow rate of auto purge exhaust (Eq. (14)) is a function of the inlet/outlet gas compositions, number of fuel cells, stack current, and the manual purge flow rate. The flow rate of fuel gas in the fuel cell system can also be determined by measuring the exhaust flow rate and the change in its composition.

3.2. Purge cell and MEA voltage in the stack

A real fuel cell system is usually assembled in a stack format consisting of a group of fuel cells. In the cells near the fuel exhaust outlet, impurities gradually accumulate needing to be well managed in order to provide enough hydrogen fuel for these particular anodes to perform the electrochemical reaction. Periodical purge or continuous exhaust release is normally required for diluting or refreshing the exhaust build-up although a stack may be claimed as a dead-end structure. The exhaust purge is normally triggered by monitoring the voltage of the purge cell portion when reaching the threshold values [7]. In consideration of the system operation and safety reasons, the Nexa™ power system was chosen for the lab test.

The purge cell voltage was monitored as a function of operation time. Its data value (Fig. 3) was sensitively related to the nitrogen gas in the anode fuel line. There is no voltage drop with a pure hydrogen feed at a manual exhaust flow rate of 1.80 ml s^{-1} . The system was then switched to a 7.3% nitrogen fuel feed. The fuel and exhaust gas management on the anode side was conducted by the system controls automatically purging exhaust if the fuel cells were operated at an unacceptably low cell voltage. After the PEFC system was completely switched to the nitrogen feed, the flow rate of the manual exhaust decreased to 0.86 ml s^{-1} . The auto purge frequency was 15.4 mHz. It was reduced to 12.5 mHz when the exhaust flow rate was increased to 1.90 ml s^{-1} (the flow rate was measured as 3.63 ml s^{-1} for a pure hydrogen feed). As shown in Fig. 4, data were also collected at different flow rates

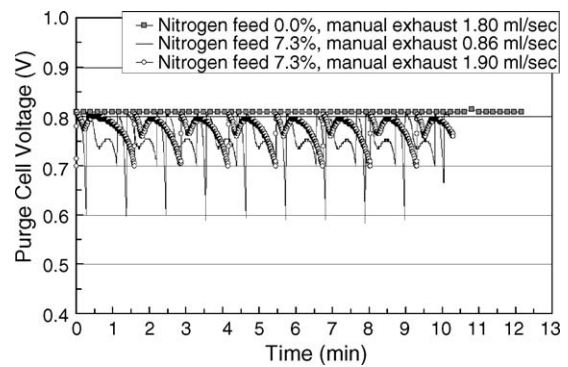


Fig. 3. Purge cell voltage as a function of operation time with ca. 7.3% nitrogen fuel feed in comparison with pure hydrogen operation. Exhaust flow rate in situ measured by the same fuel feed; stack temperature: 31 °C.

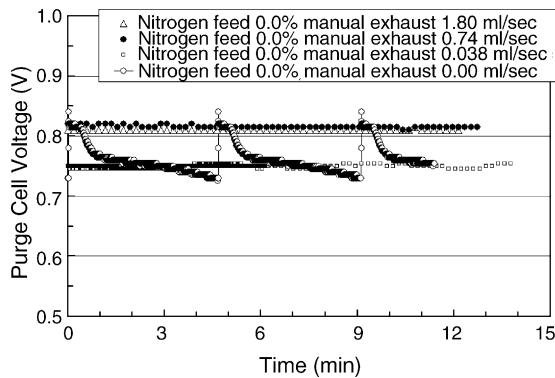


Fig. 4. Purge cell voltage as a function of operation time without inert gas feed. Exhaust flow rate measured by pure hydrogen (>99.99%) feed; stack temperature: 31 °C.

of the manual exhaust with a pure hydrogen fuel feed. During normal operation, the purge frequency (f_p) was ca. 3.7 mHz at a stack power of 200 W. With a proper manual rate increase of 0.038 ml s^{-1} , the minimum purge voltage (i.e., critical flow rate, discussed later in detail) no longer decreased and the auto purge stopped (Fig. 4). With the second rate increase of the manual purge to 0.74 ml s^{-1} , the maximum purge voltage was a steady 0.82 V. Even when the manual purge flow rate was later increased to 1.80 ml s^{-1} , the purge voltage remained in the same range of values. Small differences in voltage were caused by a negligible change of temperature. The Nexa™ system, fed by pure hydrogen, was tested at a 200 W stack power level for membrane dry-out resulting in no voltage diminution during a testing period of 48 min at an exhaust flow rate of 5 ml s^{-1} (Fig. 5). The measured purge frequency (f_p) was ca. 16.3 mHz at a stack power of 1200 W.

The fuel cell system (Nexa™ #527) was tested at 7.5% nitrogen content, balanced with pure hydrogen. The stack and control system were successfully operated for start-up and the duration of inert gas intake. The single fuel cell, i.e., MEA voltage (No.13, 33 and 47 from the anode fuel inlet to the exhaust outlet) was measured at a level of 0.70 V at a stack

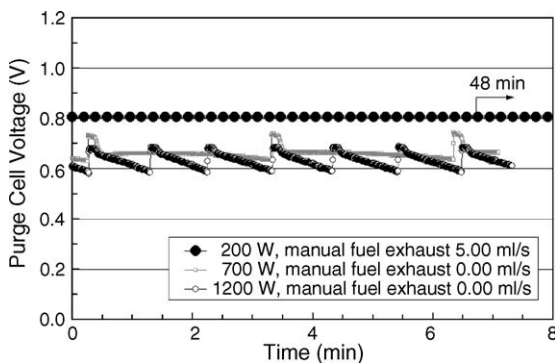


Fig. 5. Membrane dry-out and purge frequency tests using pure hydrogen fuel at different stack power levels. Exhaust flow rate measured by pure hydrogen (>99.99%) fuel feed. Stack operated at 200 W (48 min tested), ~32 °C; 700 W, ~50 °C; 1200 W ~65 °C.

power of 200 W, but the dynamic voltage signals were difficult to collect simultaneously for single fuel cells. A system from National Instruments was set up in order to obtain voltage information for MEAs at different locations of the stack [10]. The voltage across each cell can then be monitored for further diagnosis purpose at different inert gas levels.

Carbon dioxide was more difficult than nitrogen to obtain a good gas mixture in the lab because carbon dioxide (1.964 mg ml^{-1}) is 22 times heavier than hydrogen ($0.0893 \text{ mg ml}^{-1}$) at 1 atm and 0 °C (STP). The fuel cell system with an exhaust flow rate of 1.70 ml s^{-1} at 200 W stack power was successfully operated with a ca. 7% carbon dioxide feed for 50 min. Both PEFC stack tests with ca. 7% nitrogen or carbon dioxide demonstrated that the Nexa™ power module is capable of operation with higher levels of inert gases with an additional exhaust purge line.

3.3. Critical flow rate of the anode fuel exhaust

While the system was running, it was noticed that there exists a critical flow rate between the auto purge and manual purge line. If the manual exhaust flow rate is lower than the critical value, the system continually purges with automatic controls. If higher than the critical value, there is no need for the automated purge. This is potentially beneficial for fuel recycle mode design and for increasing the fuel efficiency. As shown in Fig. 6, the critical value can be approximately measured by manually adjusting the back pressure valve. The auto purge subsystem gradually stops when the manual purge flow rate is slowly increased to a certain limit, at which point, the purge cell voltage maintains a stable minimum value with no need for automatic purge.

After fuel cell system startup, the purge cell voltage reduced gradually and the anode fuel exhaust line purged for voltage restoration and performance recovery (Figs. 3 and 6). After an automatic purge occurred, the purge cell voltage increased a maximum level. This is probably caused by a temporary inert gas release at catalytic active sites during the quick purge process. The voltage rapidly increased after the

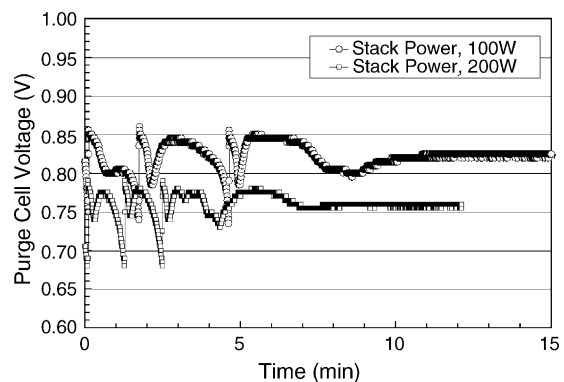


Fig. 6. Purge cell voltage as a function of operation time at two different stack power levels. Manual exhaust purge started after two purge cycles; stack temperature: 28–30 °C.

auto purge. The purge cell voltage then decreased quickly, followed by a peak of increase, and then moved to the next purge cycle. This may result from the fresh fuel gas which was purged into the anode in due form and replaced the impurities absorbed at the catalytic active sites. At this moment, the manual purge valve was slightly opened and the purge cell voltage gradually increased (Fig. 6). The flow rate was maintained at a minimum level and the purge cell voltage was also maintained as a constant. If a slight voltage change occurred, a small adjustment to the back pressure was necessary to keep this parameter constant. This minimum flow rate (v_e), or critical flow rate of the anode fuel exhaust stream, was a useful parameter for calculating the auto purge frequency. As a whole, the critical flow rate is determined by measuring the manual purge flow rate (v_h) at an auto purge flow rate (v_a) close to 0. This minimum value can be written:

$$v_{h,\min} = \lim_{v_a \rightarrow 0} = \frac{1 - X_{O1}}{X_{O1} - x_{h1}} \frac{mI}{2F} \quad (15)$$

Theoretically, the auto purge frequency can be approximately determined by using the above critical flow rate in conjunction with the released volume of gas in each auto purge. The fuel cell stack is safe for operation while its total purge flow rate is no less than this minimum flow rate. In the practical operation of the fuel cell system, the requirement for the overall auto purge flow rate is a little higher than the calculated value because the operational purge frequency (f_p) cannot be set to a value of ∞ (With the assumption of $f_p \rightarrow 0$, the critical minimum flow rate is obtained). There is also an increase in the anode concentration polarization while the impurity builds up between the purge intervals. Once the manual purge reaches its critical value, the inert gas exhaust attains its maximum operational concentration of impurity:

$$x_{h2,\max} = (1 - X_{O1}) \left(1 + \frac{mI}{2Fv_{h,\min}} \right) \quad (16)$$

at the minimum flow rate. Otherwise, the purge cell voltage, near the fuel exhaust side, decreases to an inoperable condition requiring further purge for MEA polarization and PEFC lifetime protection.

As described earlier, the Nexa™ system and an additional exhaust line were connected to measure the critical flow rate. At the manual exhaust outlet, the GC gas sample for nitrogen composition was taken after recording data of the exhaust flow rate. The manual purge flow rate was measured during steady stack operation at a specified power level. The fuel consumption rate and total exhaust flow rate in all of the stack flow channels were calculated using Eqs. (10) and (13).

The auto purge flow rate is a dynamic value; thus, an average flow rate was approximated and substituted into the data. The concentration of inert gas in the exhaust decreased with increasing the manual exhaust flow rate. The manual flow rate was measured while the auto purge was stopped ($f_p = 0$) and the voltage of the purge cell portion was constant. As shown in Table 1, the auto purge rates were close to zero, although no auto purge was actually observed during the test process. This type of operation provides a dual use of membranes in the MEAs as both gas purifiers and solid electrolytes. Optimization of the fuel and exhaust gas management potentially increases system energy efficiency. This operation reduces the system size and operational complexity by eliminating the palladium purifying system between a fuel reformer and a PEFC stack, which normally requires ~ 250 psi and $300\text{--}400^\circ\text{C}$ to obtain a hydrogen fuel pure enough ($>0.01\%$) for the PEFC system requirements under normal operating procedures.

In further lab tests, a hydrocarbon reformat containing high level of CO_2 and H_2O was also used in the Nexa™ stack after troublesome CO and H_2S poisons were removed. Because small amounts of poisonous carbon monoxide were introduced into the fuel cell system, the overall stack voltage dropped gradually (Fig. 7). The voltage of the purge cell at the end of the stack measured slightly higher than the average cell voltage in the PEFC stack. This means that a concentration gradient of carbon monoxide existed inside the fuel gas distribution channel. The front fuel cells in the stack were more likely to receive the poisons causing temporary catalyst deactivation. This phenomenon will be further examined in the diagnostic analysis of the fuel cell system with small amounts of poison in fuel gas feeds.

Table 1

Fuel gas and its exhaust gas compositions as well as different flow rates in the anode side of the system (room temperature 20°C , fuel to system 20 psig)

Stack power, P_s (W)	Stack current, I_s (A)	H_2 at inlet, X_{O1} (%)	N_2 at inlet, X_{O2} (%)	H_2 at outlet, x_{h1} (x_{e1}) (%)	N_2 at outlet, x_{h2} (x_{e2}) (%)	Bubble flow, v_h^a (ml s^{-1})	H_2 used in the cells, $mI/(2F)^b$ (ml s^{-1})	Inlet flow, u_0 (ml s^{-1})	Outlet flow, v_e (ml s^{-1})	Auto flow, v_a (ml s^{-1})
50	1.22	93.90	6.10	21.60	78.40	0.51	7.143	7.75	0.603	0.090
50	1.34	93.00	7.00	3.96	96.04	0.51	7.85	8.47	0.617	0.109
100	2.58	94.25	5.75	2.69	97.31	0.97	15.11	16.06	0.949	-0.02
100	2.58	92.50	7.50	7.70	92.30	1.16	15.11	16.45	1.336	0.176
150	4.40	94.25	5.75	31.80	62.80	1.65	23.65	25.83	2.178	0.53
200	5.64	92.50	7.50	8.41	91.59	2.22	33.02	35.96	2.95	0.725
200	5.47	97.60	2.40	14.60	85.40	0.80	32.03	32.96	0.926	0.126
200	5.47	93.90	6.10	10.20	89.80	1.85	32.03	34.36	2.334	0.484

^a The critical flow rate was approximately measured from the manual gas exhaust stream.

^b The consumption flow rate of hydrogen fuel was converted to ml s^{-1} as it was assumed the stream in the stack was at 20°C and 1 atm.

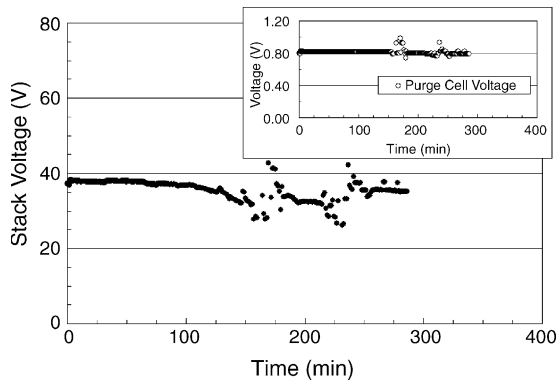


Fig. 7. Stack and purge cell voltage as a function of operation time when using a hydrocarbon reformat containing high levels of CO_2 and H_2O (hydrogen $\sim 98\%$, $\text{CO} < 100$ ppm, $\text{CO}_2 < 100$ ppm, $\text{CH}_4 \sim 2\%$, stack temperature 30°C).

3.4. Fuel cell stack efficiency and overall system efficiency

The fuel cell efficiency is the genuine efficiency of the electrochemical reaction. Thermal (energy) efficiency of the fuel cells is defined as the amount of useful energy produced relative to the change in stored chemical energy (commonly referred to as thermal energy, i.e., enthalpy change) that is released when a fuel reacts with an oxidant [11]. The ideal energy efficiency of the H_2/O_2 fuel cell ($\eta_{\text{fe}}^\circ = \Delta G^\circ / \Delta H^\circ$) is 83% when the cell is reversibly operated to produce liquid water and heat is not considered. A more general expression for the energy efficiency of the fuel cells (or stack) is summarized:

$$\eta_{\text{fe}} = \frac{-nF\bar{E}_{\text{fc}} + q_w/m}{\Delta H^\circ} = \frac{-nF\bar{E}_{\text{fc}} + q_w/m}{-nFE_{\text{fc}}^\circ/\eta_{\text{fe}}^\circ} \quad (17)$$

The fuel cell energy efficiency increases by the co-generation of heat and electricity if the formed heat (q_w) from the cooling system is partially recycled for hot water, etc. For a common PEM fuel cell where q_w is not employed for useful work ($q_w = 0$), the single cell energy efficiency is simplified:

$$\eta_{\text{fe}} = \frac{\eta_{\text{fe}}^\circ \bar{E}_{\text{fc}}}{E_{\text{fc}}^\circ} \quad (18)$$

The fuel cell (stack) efficiency in actual operation is given:

$$\eta_{\text{fc}} = \frac{\bar{E}_{\text{fc}}}{E_{\text{fc}}^\circ} = \frac{E_s}{mE_{\text{fc}}^\circ} \quad (19)$$

This expression is also called voltage efficiency [12].

Efficiencies of the PEFC stack and its system are the most significant concerns regarding the whole system performance. When the NexaTM PEFC system was operated at ca. 200 W stack power with 7.5% nitrogen feed ($v_h = 2.22 \text{ ml s}^{-1}$, Table 1), the fuel cell stack efficiency of 47 total cells was approximately 64% (stack voltage, $E_s = 35.95 \text{ V}$; theoretical cell voltage, $E_{\text{fc}}^\circ \approx 1.2 \text{ V}$). Due to the power consumed by the electronic controls and air compressor, the stack

output efficiency for useful work was 75%. While nitrogen in the anode stream was concentrated from 7.5% to 91.6%, the fuel efficiency was still 99% with a manual exhaust flow rate of 2.22 ml s^{-1} and without auto purge. The overall efficiency (the energy efficiency of the whole fuel cell system) is calculated:

$$\eta_{\text{oe}} = \eta_{\text{fu}} \left(\frac{\eta_{\text{fe}}^\circ E_{\text{fc}}}{E_{\text{fc}}^\circ} \right) \eta_{\text{so}} \approx 0.394 \quad (20)$$

where the fuel gas contained 7.5% nitrogen and the PEFC was operated at its critical flow rate. The handbook [11] pointed out that the operation at higher fuel cell efficiency ($>50\%$) increases the capital cost but decreases the fuel cost. Further work is continued on the optimization of various efficiencies in order to rigorously understand the benefits of the operational method.

4. Conclusions

The PEM stack in the Ballard NexaTM system was fed with mixtures of hydrogen fuel and high levels of inert gases. After running the stack for a period of time, the purge cell voltage reduced gradually, requiring the anode fuel exhaust line to be purged for voltage restoration and performance recovery. The critical flow rate in the anode exhaust stream was obtained at a specified power level with a mass balance. Using this method of fuel gas and exhaust management, the flow rate of fuel gas in the fuel cell system was determined by measuring the exhaust flow rate and the change in its gas composition. This process provided a dual use of membranes in the MEAs as both gas purifiers and solid electrolytes. This operational method eliminated the need for a high temperature palladium separator and/or gas purifier. It also reduced the auto purge frequency required for stack performance recovery. The NexaTM PEM fuel cell system successfully utilized hydrogen feeds with ca. 7% inert gases. The fuel cell stack was safely operated with its total purge flow rate at no less than this critical flow rate. Nitrogen in the anode stream was concentrated from 7.5% fuel feed up to 91.6% N_2 exhaust; thus, this operational method still maintained a fuel efficiency of 99%. Further optimization of the fuel gas and exhaust gas management potentially increases the system energy efficiency.

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References

- [1] S. Gottesfeld, T.A. Zawodzinski, in: R.C. Alkire, H. Gerischer, D.M. Kolb, C.W. Tobias (Eds.), *Advances in Electrochemical Science and Engineering*, 5, John Wiley & Sons, Inc., New York, NY, 1997, p. 197.
- [2] F.C. Chen, R.J. Fiskum, in: *International Joint Power Generation Conference*, Atlanta, GA, 2003, pp. 302–312.
- [3] J.V.C. Vargas, J.C. Ordonez, A. Bejan, *Int. J. Heat Mass Transfer* 47 (2004) 4177–4193.
- [4] G. Boehm, D.P. Wilkinson, S. Knights, R. Schamm, N.J. Fletcher, *PCT Int. Appl.*, WO 2001043216 (2001).
- [5] W. Preidel, *PCT Int. Appl.*, WO 2003052851 (2003).
- [6] B. Wells, T.D. Uong, C.J. Schembri, G.A. James, G.A. Skinner, B. Giesecke, *PCT Int. Appl.*, WO 2003010842 (2003).
- [7] R.H. Barton, T.D. Uong, C.J. Schembri, G.A. Skinner, *PCT Int. Appl.*, WO 2003010845 (2003).
- [8] S. Ahmed, J. Kopasz, R. Kumar, M. Krumpelt, *J. Power Sources* 112 (2002) 519–530.
- [9] V. Marshall, J. Kelland, P. Grosjean, D. Bloomfield, *Proceedings of the 36th Power Sources Conference*, Cherry Hill, New Jersey, 1994, pp. 83–86.
- [10] R.U. Payne, W.H. Zhu, B.J. Tatarchuk, *Proceedings of the AIChE Annual Meeting: Fuel Cell Technology II*, Austin, Texas, 2004, p. 14h.
- [11] EG&G Technical Services Inc. and Science Applications International Corp., *Fuel Cell Handbook*, 6th ed., U.S. Department of Energy, West Virginia, 2002, p. 7 (Chapter 2).
- [12] B.L. Yi, *Fuel Cells – Principles, Techniques and Applications*, Chemical Industry Press, Beijing, 2003, p. 58.