

# Operation of PEM fuel cells at 120–150 °C to improve CO tolerance

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Received 17 September 2005; received in revised form 22 November 2005; accepted 23 November 2005

Available online 6 January 2006

## Abstract

Sulfonic acid modified perfluorocarbon polymer proton exchange membrane (PEM) fuel cells operated at elevated temperatures (120–150 °C) can greatly alleviate CO poisoning on anode catalysts. However, fuel cells with these PEMs operated at elevated temperature and atmospheric pressure typically experience low relative humidity (RH) and thus have increased membrane and electrode resistance. To operate PEM fuel cells at elevated temperature and high RH, work is needed to pressurize the anode and cathode reactant gases, thereby decreasing the efficiency of the PEM fuel cell system. A liquid-fed hydrocarbon-fuel processor can produce reformed gas at high pressure and high relative humidity without gas compression. If the anode is fed with this high-pressure, high-relative humidity stream, the water in the anode compartment will transport through the membrane and into the ambient pressure cathode structure, decreasing the cell resistance. This work studied the effect of anode pressurization on the cell resistance and performance using an ambient pressure cathode. The results show that high RH from anode pressurization at both 120 and 150 °C can decrease the membrane resistance and therefore increase the cell voltage. A cell running at 150 °C obtains a cell voltage of 0.43 V at 400 mA cm<sup>-2</sup> even with 1% CO in H<sub>2</sub>. The results presented here provide a concept for the application of a coupled steam reformer and PEM fuel cell system that can operate at 150 °C with reformat and an atmospheric air cathode.

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**Keywords:** PEM fuel cell; Elevated temperature; Relative humidity; Fuel processor–fuel cell system; CO tolerance

## 1. Introduction

A proton exchange membrane (PEM) fuel cell converts chemical energy to electrical energy using a solid ionomer (such as Nafion<sup>®</sup>) as the electrolyte. Usually, PEM fuel cells are operated at low temperatures (<80 °C) and 100%RH. However, PEM fuel cells operated at elevated temperatures (>100 °C, atmospheric pressure and therefore dry conditions) have significant advantages over low temperature PEM fuel cells as CO poisoning at the anode is effectively alleviated [1,2]. A quantitative analysis of the free energy for H<sub>2</sub> and CO adsorption suggests that CO tolerance at the anode increases by a factor of 20, by elevating the operating temperature to higher than 100 °C [1]. CO tolerance is affected not only by the cell temperature, but by the relative humidity (RH) of reactant gases [3,4]. Higher temperature also accelerates reaction kinetics, improves heat rejection, provides higher quality waste heat, and permits

easier water management. However, at elevated temperatures and ambient pressure, the PEM fuel cell must be operated at low RH, which dries out the membrane and electrodes, resulting in significant membrane and electrode resistance. At elevated temperature, PEM fuel cells can also be operated at high RH as long as the fuel cell system can be pressurized. However, pressurization of the fuel cell system requires extra pump work, which reduces the total efficiency of the fuel cell system.

In practice, the hydrogen for a fuel cell is frequently obtained from the processing of hydrocarbon fuels by way of steam reforming. Considerable efforts have been made to integrate a fuel processor with the fuel cell in residential and automobile applications [5]. The fuel processor is usually operated at a very high temperature and pressure (up to 1000 °C and 30 atm) [6,7]. The reformed gases, including hydrogen, carbon dioxide, carbon monoxide, water, etc., are at this high pressure before they enter the anode of the fuel cell system. Therefore, it is possible to run a fuel cell with the anode pressurized and the cathode at ambient pressure without lowering the system efficiency. Pressurization of the anode makes it possible to increase the anode RH while

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running a fuel cell at elevated temperature. The increased RH at the anode results in more water at the anode side. Due to the concentration gradient of water between the anode and the cathode, the water in the anode side transports through the membrane and reaches the cathode, wetting the membrane and the cathode.

This study investigated anode pressurization for hydrogen and hydrogen–carbon monoxide/atmospheric air cathode systems. The effects of anode pressurization on membrane resistance, cell voltages, hydrogen crossover and electrochemical area (ECA) were evaluated. These results provide a concept for the application of a coupled steam reformer and PEM fuel cell system that can operate at 150 °C with reformat and an atmospheric air cathode.

## 2. Experimental

### 2.1. Membrane and electrode assembly (MEA)

The membrane electrode assemblies were prepared by applying both the anode and the cathode catalyst layers directly onto a composite Nafion®–Teflon® phosphotungstic acid (NTPA) membrane, with a thickness of 25–50 μm. The NTPA membrane was made by mixing 1.5 ± 0.01 g PTA power (Aldrich Chemical Corp., Milwaukee, WI) with 120 ± 0.1 g 5% Nafion® solution (Solution Technologies, Mendenhall, PA) and then impregnating the mixed solution onto 18 μm porous Teflon® film (Tetratex Products Inc., Camarillo, CA) [8]. The cathode catalyst, 46.6 wt.% Pt/C (Tanaka Kikinokogyo, Tokyo, Japan), was mixed with 5% Nafion® solution by stirring and ultrasonic dispersion before it was applied onto the membrane. The cathode loading of precious metal was 0.25 mg cm<sup>-2</sup> and the Nafion® content was 35 wt.%. On the anode, Pt–Ru black (Alfa Aesar, Ward Hill, MA) was applied onto the membrane with 1.0 mg cm<sup>-2</sup> catalyst loading and 10 wt.% Nafion® content. The catalyst-coated membrane was sandwiched between two SGL gas diffusion layers (SGL Carbon Group, Wiesbaden, Germany) to obtain a 5 cm<sup>2</sup> MEA for single-cell polarization measurements. A single serpentine 5 cm<sup>2</sup> active area graphite flow field was used (ElectroChem Inc., Woburn, MA) for cell assembly. On both anode and cathode sides, a 300 μm thick gasket made of Teflon® film (MSC industrial supply, Co. Inc., Melville, New York) was used. A 2.8 N·m torque was applied to seal the gaskets.

### 2.2. Linear sweep voltammetry and cyclic voltammetry

Linear sweep voltammetry and cyclic voltammetry (CV) were performed using a PAR 273A Potentiostat (Princeton Applied Research, Oak Ridge, TN) with hydrogen at the anode and nitrogen at the cathode first at 80/80/80 °C (100%RH, cell temperature/anode saturator temperature/cathode saturator temperature) and then 120 and 150 °C with different RHs. The RHs of anode and cathode gases were controlled by saturators in a MEDUSA™ test station (Teledyne Energy Systems Inc., Los Angeles, CA). Before the measurement, the MEA was equilibrated at the experimental condition for 1 h. Electrochemical

area was calculated based on the following equation:

$$ECA = \frac{Q_H}{[Pt] \times 0.21}$$

Here [Pt] represents the platinum loadings (mg cm<sup>-2</sup>) in the cathode,  $Q_H$  the charge for hydrogen adsorption (mC cm<sup>-2</sup>), which can be calculated from the CVs based on the integration of hydrogen adsorption area, and 0.21 in mC cm<sup>-2</sup> platinum represents the charge required to oxidize a monolayer of hydrogen on bright Pt [9].

### 2.3. Cell voltages and resistances

A 890C load box (Scribner Associates Inc., Southern Pines, NC) was used to measure cell voltage and resistance. Five minutes were spent at each current density with the cell voltage collected every 20 s. Cell internal resistance,  $R$ , was measured and recorded at current densities higher than 100 mA cm<sup>-2</sup> using the current interrupt technique with the load box and Fuel Cell V3.2 software (Scribner Associates Inc., Southern Pines, NC). IR-corrected cell voltages are equal to the cell voltages plus  $iR$ . The utilization of hydrogen was 33% for current densities above 1000 mA cm<sup>-2</sup> whereas a fixed flow rate of 0.1 L min<sup>-1</sup> was used below this current. The utilization of oxygen in air was 25% for current densities above 300 mA cm<sup>-2</sup> whereas a fixed flow rate of 0.1 L min<sup>-1</sup> was used below this current.

## 3. Results and discussions

### 3.1. Cell resistance and voltages at ambient pressure

Fig. 1 shows the cell voltages and resistances using H<sub>2</sub>/air at two temperatures, 80 and 120 °C, and ambient pressure. The thickness of the NTPA membrane was 24 μm. The first condition, 80/80/80 °C (cell/anode saturator/cathode saturator temperature), is frequently used for low temperature PEM fuel cells. It corresponds to 100%RH, with a 0.53 atm air partial pressure, 0.53 atm hydrogen partial pressure and 0.47 atm water partial pressure. The second condition, 120/90/90 °C, has a relative humidity of 35% at the inlet with a 0.3 atm air partial pressure, 0.3 atm hydrogen partial pressure and 0.7 atm water partial pressure. At ambient pressure, this is close to the highest RH that can be obtained for a cell operated at 120 °C without severe performance loss due to low reactant partial pressures. The cell voltage and resistance at a current density of 400 mA cm<sup>-2</sup> has been selected to make comparisons. At 80 °C and 100%RH, the membrane resistance is 0.07 Ω·cm<sup>2</sup>, and the cell voltage is 0.7 V at 400 mA cm<sup>-2</sup>. At 120 °C and 35%RH, the membrane resistance is 0.21 Ω·cm<sup>2</sup> and the cell voltage is 0.56 V. The low cell voltage at 120 °C/35%RH/35%RH is due in large part to the high membrane resistance at low RH. The higher cell voltage at the 80 °C condition is also due to higher hydrogen and air partial pressures (0.53 atm) than those at the 120 °C condition (0.3 atm). The operation at 120 °C/35%RH/35%RH is used here as the base-line condition. Cell voltage and resistance with anode pressurization at

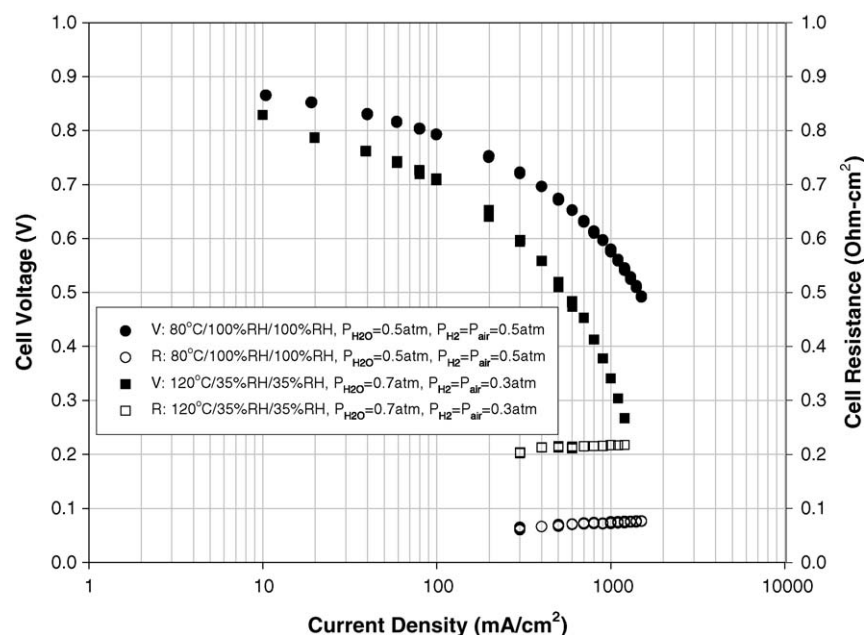


Fig. 1. Cell voltages and resistances at 80 and 120 °C without pressurization. 80 °C/100%RH/100%RH stands for cell temperature/anode RH/cathode RH, same as below. NTPA membrane: 24  $\mu\text{m}$ . Anode:  $\text{H}_2$ , stoic 3; cathode: air, stoic 4.

both 120 and 150 °C will be compared to those at this base-line condition.

### 3.2. Anode pressurization at 120 °C

To increase the RH of the MEA, the total pressure of the anode was increased to 1.7 atm and the anode saturator temperature was elevated to 110 °C, but the cathode saturator was still kept at 90 °C and the cathode pressure was kept at 1 atm. The thickness of the NTPA membrane used here was 36  $\mu\text{m}$ . The condition, as represented by 120/110/90 °C, has 72%RH at the anode and 35%RH at the cathode (120 °C/72%RH/35%RH). The water partial pressure at the anode is 1.4 atm and the hydrogen partial pressure is 0.3 atm. At the cathode, the water partial pressure is still 0.7 atm and the air pressure is 0.3 atm. Fig. 2 shows the cell voltages and resistances at 120 °C/72%RH/35%RH, as compared to the base line 120 °C/35%RH/35%RH. At 120 °C/72%RH/35%RH, the membrane resistance is 0.16  $\Omega\text{-cm}^2$ , which is half of the resistance at 120 °C/35%RH/35%RH, 0.33  $\Omega\text{-cm}^2$ . The resistance here at 120 °C/35%RH/35%RH is higher than the 0.21  $\Omega\text{-cm}^2$  of Fig. 1 because the membrane thickness for this cell is greater (36  $\mu\text{m}$  compared to 24  $\mu\text{m}$ ). A thicker membrane was used to lower the hydrogen crossover due to the pressure gradient across the membrane. Corresponding to this significantly reduced membrane resistance, the cell voltage at 120 °C/72%RH/35%RH increases to 0.66 V, compared to 0.56 V at 120 °C/35%RH/35%RH. The high RH (72%) at the anode introduces a large amount of water into the anode, while the cathode side has much less water since its RH is only 35%RH. Due to the water concentration gradient and pressure gradient across the membrane from the anode to the cathode, the water molecules move from the anode to the cathode. The transported water from the anode efficiently wets the membrane

and the cathode, reducing the cell resistance. There are different mechanisms by which water can transport from the anode to the cathode, such as convection, diffusion and electroosmotic drag.

### 3.3. Anode pressurization at 150 °C

Fig. 3 shows results using anode pressurization of the same cell to 2.7 atm at 150 °C. The condition, 150/126/90 °C, has a 50%RH at the anode and 15%RH at the cathode (150 °C/50%RH/15%RH). The water partial pressure at the anode is 2.4 atm and the hydrogen partial pressure is 0.3 atm. At the cathode, the water partial pressure is still 0.7 atm and the air pressure is 0.3 atm. The membrane resistance at 150 °C/50%RH/15%RH is 0.28  $\Omega\text{-cm}^2$ , which is also lower than 0.33  $\Omega\text{-cm}^2$  at the baseline condition 120 °C/35%RH/35%RH. However, the cell voltage at 150 °C/50%RH/15%RH and 400  $\text{mA cm}^{-2}$  is 0.44 V, which is much lower than the 0.56 V at 120 °C/35%RH/35%RH. Although anode pressurization at 150 °C reduces the membrane resistance to below that of 120 °C without pressurization, the cell voltage decreases 120 mV. This first may be explained by the decrease in open circuit voltage (OCV). According to the Nernst equation, the higher temperature and the increased water partial pressure lower the OCV. At the 150/126/90 °C (150 °C/50%RH/15%RH) condition, not only is the temperature higher than that at 120 °C, but the water partial pressure at the anode, 2.4 atm, is also much larger than at 120/90/90 °C (120 °C/35%RH/35%RH), 0.7 atm. At the 150/126/90 °C (150 °C/50%RH/15%RH), the actual OCV is only 0.86 V, which is lower than the 0.91 V at 120/90/90 °C (120 °C/35%RH/35%RH). However, the OCV only results in a 50 mV decrease and of the total 120 V cell voltage decrease. The lower performance at 150 °C/50%RH/15%RH may also come

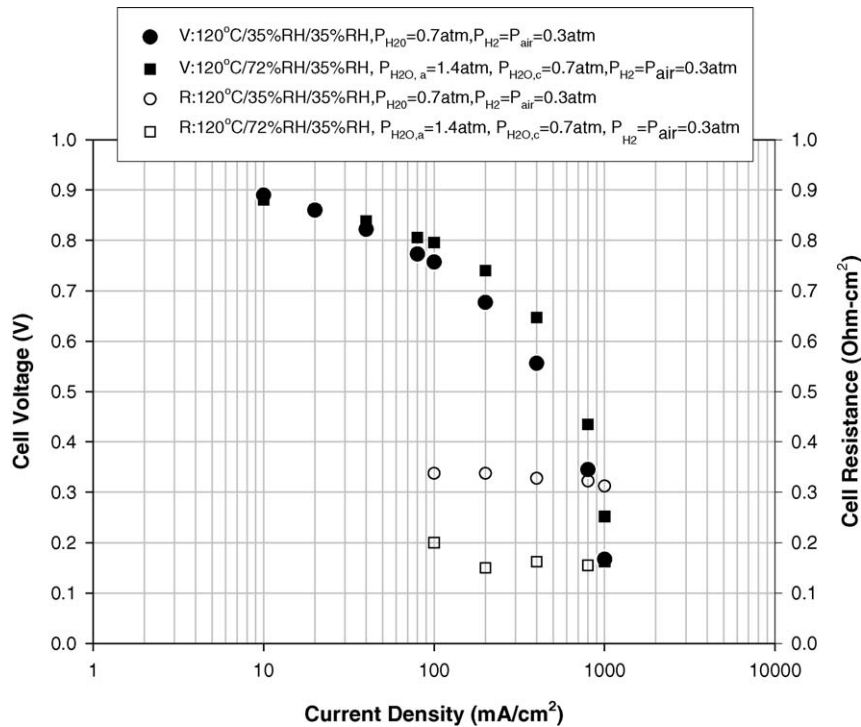


Fig. 2. Anode pressurization of PEM fuel cell at 120 °C. NTPA membrane: 36  $\mu\text{m}$ . Anode:  $\text{H}_2$ , stoic 3; cathode: air, stoic 4. 120 °C/35%RH/35%RH:  $P_{\text{total}} = 1 \text{ atm}$ ,  $P_{\text{H}_2\text{O}} = 0.7 \text{ atm}$ ,  $P_{\text{H}_2} = P_{\text{air}} = 0.3 \text{ atm}$ ; 120 °C/72%RH/35%RH:  $P_{\text{total, anode}} = 1.7 \text{ atm}$ ,  $P_{\text{H}_2\text{O, anode}} = 1.4 \text{ atm}$ ,  $P_{\text{H}_2} = 0.3 \text{ atm}$ .

from a decrease in the effective surface area of the electrodes, or an increase in the ionic electrode resistance of the cathode under these harsh conditions. To study the interface and electrode condition, linear sweep voltammetry and cyclic voltammetry

were conducted, which determined the hydrogen crossover of the NTPA membrane and the ECA of the cathode.

Table 1 shows the hydrogen crossover and partial and total pressures at four conditions: 80 °C/100%RH/100%RH,

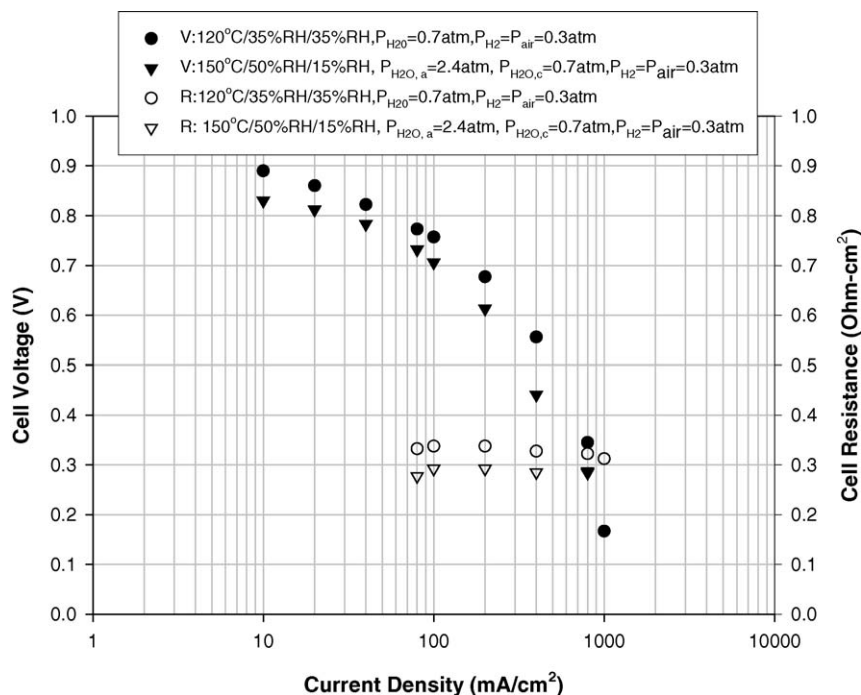


Fig. 3. Anode pressurization of PEM fuel cell at 150 °C. NTPA membrane: 36  $\mu\text{m}$ . Anode:  $\text{H}_2$ , stoic 3; cathode: air, stoic 4. 120 °C/72%RH/35%RH:  $P_{\text{total}} = 1 \text{ atm}$ ,  $P_{\text{H}_2\text{O}} = 0.7 \text{ atm}$ ,  $P_{\text{H}_2} = P_{\text{air}} = 0.3 \text{ atm}$ ; 150 °C/50%RH/15%RH:  $P_{\text{total, anode}} = 2.7 \text{ atm}$ ,  $P_{\text{H}_2\text{O, anode}} = 2.4 \text{ atm}$ ,  $P_{\text{H}_2} = 0.3 \text{ atm}$ .

Table 1

Hydrogen crossover at different conditions. 80/80/80 °C stands for cell temperature/anode saturator temperature/cathode saturator temperature, same for others

Conditions	80/80/80 °C, A: 100%RH, C: 100%RH	120/90/90 °C, A: 35%RH, C: 35%RH	120/110/90 °C, A: 72%RH, C: 35%RH	150/126/90 °C, A: 50%RH, C: 15%RH
<b>Cathode</b>				
$P_{\text{H}_2\text{O}}$ (atm)	0.47	0.7	0.7	0.7
$P_{\text{air}}$ (atm)	0.53	0.3	0.3	0.3
$P_{\text{Total}}$ (atm)	1.0	1.0	1.0	1.0
RH (%)	100	35	35	35
<b>Anode</b>				
$P_{\text{H}_2\text{O}}$ (atm)	0.47	0.7	1.4	2.4
$P_{\text{H}_2}$ (atm)	0.53	0.3	0.3	0.3
$P_{\text{total}}$ (atm)	1.0	1.0	1.7	2.7
RH (%)	100	35	72	50
Crossover ( $\text{mA cm}^{-2}$ )	1.9	3.1	2.1	2.6
ECA ( $\text{m}^2 \text{g}^{-1}$ )	75	63	69	39
Resistance ( $\Omega\text{-cm}^2$ )	0.065	0.32	0.16	0.28

120 °C/35%RH/35%RH, 120 °C/72%RH/35%RH and 150 °C/50%RH/15%RH. At 80 °C/100%RH/100%RH, the crossover is  $2 \text{ mA cm}^{-2}$ ; at 120 °C/35%RH/35%RH, the crossover is  $3.1 \text{ mA cm}^{-2}$ ; at 120 °C/72%RH/35%RH, however, the crossover is  $2.1 \text{ mA cm}^{-2}$ ; and at 150 °C/50%RH/15%RH, the crossover is  $2.6 \text{ mA cm}^{-2}$ . An increase of the cell temperature should increase the hydrogen crossover, as the effective diffusion coefficient of hydrogen in the membrane increases with temperature. However, the crossover at

150 °C/50%RH/15%RH and 120 °C/72%RH/35%RH is lower than that at 120 °C/35%RH/35%RH. Since the hydrogen crossover current was only 2–3  $\text{mA cm}^{-2}$  and did not linearly increase with the voltage, no electronic shorts were present in the membrane even at 150 °C/50%RH/15%RH, where the total pressure at the anode, 2.7 atm, was much higher than that at the cathode, 1.0 atm.

Fig. 4 shows cyclic voltammetry curves at different temperatures and RHs. The CV curves in the range of 0.1–0.4 V rep-

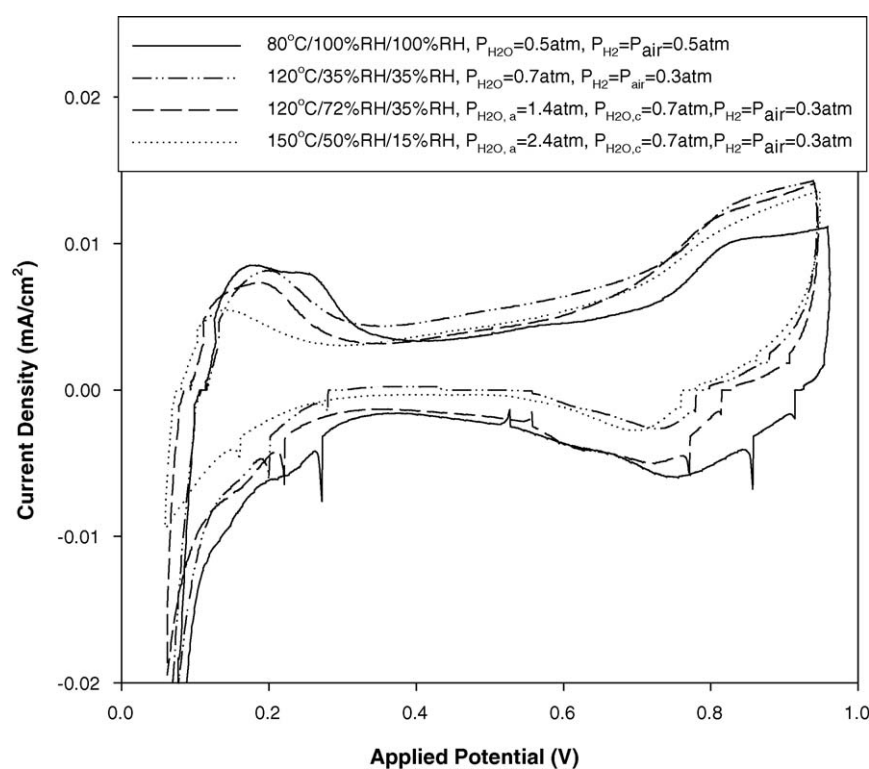


Fig. 4. Cyclic voltammetry at different temperature and relative humidity. Anode:  $\text{H}_2$ , cathode:  $\text{N}_2$ , scan rate:  $20 \text{ mV s}^{-1}$ . 120 °C/35%RH/35%RH:  $P_{\text{total}} = 1 \text{ atm}$ ,  $P_{\text{H}_2\text{O}} = 0.7 \text{ atm}$ ,  $P_{\text{H}_2} = P_{\text{air}} = 0.3 \text{ atm}$ ; 120 °C/72%RH/35%RH:  $P_{\text{total, anode}} = 1.7 \text{ atm}$ ,  $P_{\text{H}_2\text{O, anode}} = 1.4 \text{ atm}$ ,  $P_{\text{H}_2} = 0.3 \text{ atm}$ ; 150/126/90 °C:  $P_{\text{total, anode}} = 2.7 \text{ atm}$ ,  $P_{\text{H}_2\text{O, anode}} = 2.4 \text{ atm}$ ,  $P_{\text{H}_2} = 0.3 \text{ atm}$ .



resent hydrogen adsorption (negative currents) and desorption (positive currents), which can be used to calculate the electrochemical area. The three conditions,  $80^{\circ}\text{C}/100\%RH/100\%RH$ ,  $120^{\circ}\text{C}/35\%RH/35\%RH$ , and  $120^{\circ}\text{C}/72\%RH/35\%RH$ , display similar curves in the range of 0.1–0.4 V. The calculated ECAs for these three conditions are very close to each other,  $\sim 68\text{ m}^2\text{ g}^{-1}$ . However, the CV for  $150^{\circ}\text{C}/50\%RH/15\%RH$  lies much below the CV curves for the other three conditions. The calculated ECA for  $150^{\circ}\text{C}/50\%RH/15\%RH$  is only  $39\text{ m}^2\text{ g}^{-1}$ . The low ECA at  $150^{\circ}\text{C}/50\%RH/15\%RH$  may be due to a poor electrode/membrane interface at the higher temperature and lower RH at the cathode. This may explain why, at  $150^{\circ}\text{C}/50\%RH/15\%RH$ , although the membrane resistance is lower than that at  $120^{\circ}\text{C}/72\%RH/35\%RH$ , the cell voltage is also lower. Although water molecules in the anode moved to the cathode across the membrane, they might not penetrate the whole thickness of the cathode, so part of the cathode could still be at 15%RH. This dehydration of the cathode at the harsh conditions of  $150^{\circ}\text{C}/50\%RH/15\%RH$  also results in significant ionic resistance of the cathode and cathode polarization. Our previous research [10] showed that catalytic activity of the oxygen reduction reaction was strongly influenced by the RH; when the RH was below 60–70%RH, the catalytic activity decreased significantly as the RH was reduced. Our recent results have shown that the ionic resistance of the cathode may become pronounced at low RH [11]. For a uniform 20%RH in the cathode the cathode ionic resistance reaches  $0.15\ \Omega\text{-cm}^2$ , which results in about 60 mV voltage loss at  $400\text{ mA cm}^{-2}$ .

### 3.4. CO tolerance at anode pressurization

Two conditions of anode pressurization,  $120/110/90^{\circ}\text{C}$  ( $120^{\circ}\text{C}/72\%RH/35\%RH$ ), and  $150/110/90^{\circ}\text{C}$  ( $150^{\circ}\text{C}/50\%$

$RH/15\%RH$ ), were tested for CO tolerance at a concentration of 0.1% and 1%. Fig. 5 shows the CO tolerance at  $120^{\circ}\text{C}/72\%RH/35\%RH$ . There are three cell voltage curves in the figure. For pure  $\text{H}_2$ , the cell voltage is 0.66 V at  $400\text{ mA cm}^{-2}$ . For 0.1% CO, the cell voltage is reduced to 0.52 V at  $400\text{ mA cm}^{-2}$ . Finally for 1% CO, the cell voltage decreases drastically to 0.05 V at  $400\text{ mA cm}^{-2}$ . At the base line  $120^{\circ}\text{C}/35\%RH/35\%RH$  with pure hydrogen at ambient pressure on both sides, the cell voltage was 0.56 V at  $400\text{ mA cm}^{-2}$ . Therefore, for a CO concentration such as 0.1%, anode pressurization generates a cell voltage close to that at  $120^{\circ}\text{C}/35\%RH/35\%RH$  under pure hydrogen (0.52 V versus 0.56 V). This is mainly due to decreased membrane resistance at higher RH resulting from the anode pressurization. Increased RH may also improve the CO tolerance itself due to easier CO oxidation on the surface of platinum at higher RH [4]. Fig. 5 also shows the resistance for pure hydrogen and different concentrations of CO. The membrane resistance at 0.1% and 1% CO is slightly higher than that for pure hydrogen. That is  $0.15\ \Omega\text{-cm}^2$  for pure hydrogen,  $0.18\ \Omega\text{-cm}^2$  for 0.1% CO, and  $0.21\ \Omega\text{-cm}^2$  for 1% CO. This difference may be because, for pure hydrogen, most of the anode reaction occurs at the anode interface near the membrane since only a small fraction of the anode catalyst is needed; under such circumstances, any anode polarization would not contribute to the cell resistance determined using the current interruption technique even if the anode has a slight internal electronic resistance. This occurs because the ionic resistance in the anode is small due to the short distance the ions must migrate and the anode has considerable double-layer capacitance. At higher CO concentrations, the whole depth of the catalyst structure is used because of the slower reaction kinetics. Then, with a significant ionic resistance, part of the anode polarization will be included in the cell resistance deter-

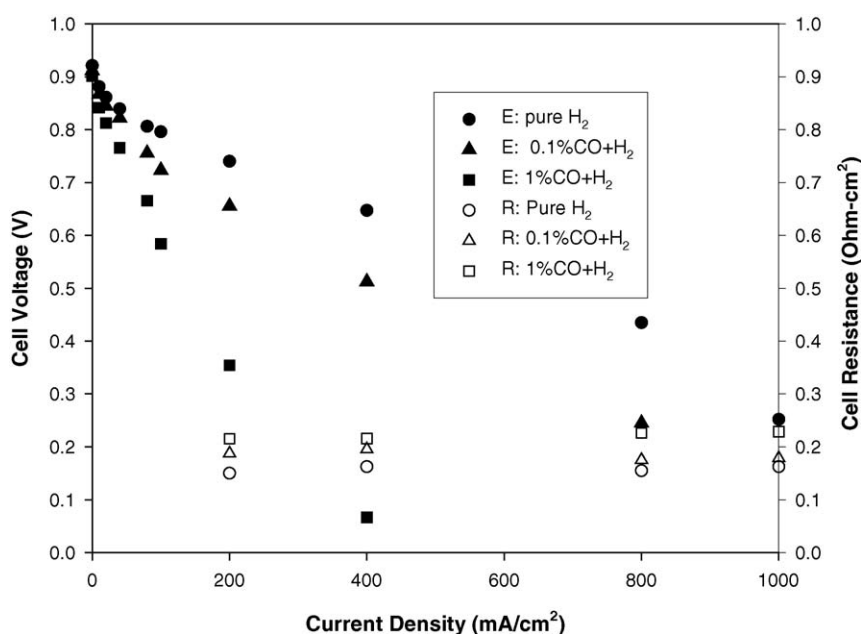


Fig. 5. CO tolerance at  $120^{\circ}\text{C}/72\%RH/35\%RH$  with anode pressurization.  $P_{\text{total, anode}} = 1.7\text{ atm}$ ,  $P_{\text{H}_2} = 0.3\text{ atm}$ ,  $P_{\text{H}_2\text{O, anode}} = 1.4\text{ atm}$ ;  $P_{\text{total, cathode}} = 1.0\text{ atm}$ ,  $P_{\text{air}} = 0.3\text{ atm}$ ,  $P_{\text{H}_2\text{O, anode}} = 0.7\text{ atm}$ . NTPA membrane:  $36\ \mu\text{m}$ . Anode: stoic 3; cathode: air, stoic 4.

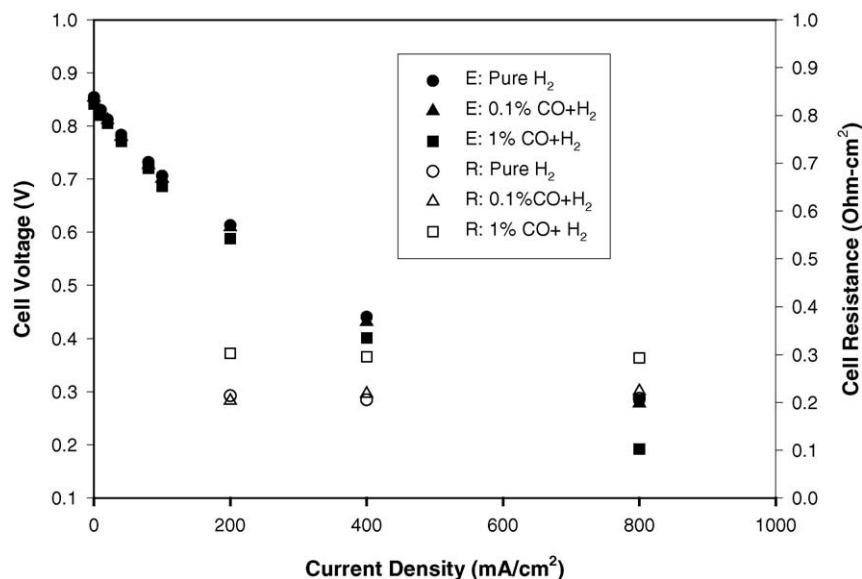


Fig. 6. CO tolerance at 150 °C/50%RH/15%RH with anode pressurization.  $P_{\text{total, anode}} = 2.7$  atm,  $P_{\text{H}_2} = 0.3$  atm,  $P_{\text{H}_2\text{O, anode}} = 2.4$  atm;  $P_{\text{total, cathode}} = 1.0$  atm,  $P_{\text{air}} = 0.3$  atm,  $P_{\text{H}_2\text{O, cathode}} = 0.7$  atm. NTPA membrane: 36  $\mu\text{m}$ . Anode: stoic 3; cathode: air, stoic 4.

mined by current interruption if some electronic resistance is present in the anode. This effect becomes more pronounced at lower RH when the ionic conductivity of the electrolyte is reduced.

Fig. 6 shows the CO tolerance at 150 °C/50%RH/15%RH. The effect of increased temperature on alleviating CO poisoning is very obvious in this case. The three curves: pure hydrogen, 0.1% and 1% CO almost coincide. For pure H<sub>2</sub>, the cell voltage is 0.48 V at 400 mA cm<sup>-2</sup>; for 0.1% CO, the cell voltage is 0.47 V at 400 mA cm<sup>-2</sup>. Even at 1% CO concentration, the cell voltage is still 0.43 V at 400 mA cm<sup>-2</sup>. The increased CO concentration at 150 °C does not poison the catalyst as much as at 120 °C. The resistance also increases with the CO concentration, that is, 0.21  $\Omega\text{-cm}^2$  for pure hydrogen, 0.23  $\Omega\text{-cm}^2$  for 0.1% CO, and 0.29  $\Omega\text{-cm}^2$  for 1% CO. Cell performance with 1% CO at 150 °C/50%RH/15%RH should be further improved by optimizing both the cathode and anode. In this study, the anode catalyst was a Pt–Ru black catalyst and the cathode catalyst was carbon-supported platinum. An additional way to improve performance would be to use carbon-supported Pt–Ru at the anode and Pt–Co at the cathode.

#### 4. Conclusions

This research provides a framework for the application of a coupled steam reformer and PEM fuel cell system that can operate at 120 or 150 °C with reformed gas and an atmospheric air cathode.

At 120/110/90 °C (120 °C/72%RH/35%RH), anode pressurization (1.7 atm) brings significant reduction of cell resistance and results in 90 mV cell voltage gain at 400 mA cm<sup>-2</sup>, compared to 120/90/90 °C (120 °C/35%RH/35%RH) without pressurization (1 atm). At 150/126/90 °C (150 °C/50%RH/15%RH), pressurization (2.7 atm) decreases cell resistance, but does

not increase cell voltage. Cyclic voltammetry indicates that at 150 °C/50%RH/15%RH, the ECA is much lower than at 120 °C/72%RH/35%RH and 120 °C/35%RH/35%RH. The operation of PEM fuel cells at 150 °C/50%RH/15%RH may generate high ionic and electronic resistance at the cathode and cause serious cathode polarization. Anode pressurization makes it possible to operate PEM fuel cells at higher CO levels. At 120 °C/72%RH/35%RH, 0.56 V cell voltage at 400 mA cm<sup>-2</sup> is obtained for 0.1% CO in H<sub>2</sub>; at 150 °C/50%RH/15%RH, 0.43 V at 400 mA cm<sup>-2</sup> is obtained for 1% CO in H<sub>2</sub>. The advantage of anode pressurization was demonstrated, but the conditions selected to conduct this research are different from actual reformed gases. At 150 °C/50%RH/15%RH, the water partial pressure at the anode is 2.4 atm and the hydrogen and CO pressure is 0.3 atm. In an actual steam reforming system, the anode composition is more likely to be near 38% H<sub>2</sub>, 16% N<sub>2</sub>, 10% CO, 6% CO<sub>2</sub> and 30% H<sub>2</sub>O. In this case, the total pressure of reformed gases must be near 10 atm, to obtain 3 atm of water vapor. In such a situation the large pressure difference between anode and cathode is a concern with respect to membrane strength. Also, water shifting conversion is needed to bring down the CO content in the mixed gases.

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