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An experimental study on the hydrogen crossover in polymer electrolyte membrane fuel cells for various current densities

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HIGHLIGHTS

- Hydrogen crossover rate are measured according to diverse current densities.
- The higher hydrogen crossover rate is detected for the elevated clamping pressure.
- The higher hydrogen crossover rate is detected for the elevated relative humidity.
- Hydrogen concentration is almost constant under various current densities.
- Hydrogen crossover rate is gradually increased as the current density is increased.

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ABSTRACT

The effect of the clamping pressure and the relative humidity on gas crossover is studied in this research. It is central to this paper that the effect of the operating parameters on the hydrogen crossover is analyzed for various current densities. The performance and the hydrogen crossover rate are measured simultaneously according to the different clamping pressure and relative humidity at each current density. The effect of both parameters on the system performance coincides well with other researches. The higher hydrogen concentration and crossover rate are detected for the elevated clamping pressure and relative humidity. Interesting result from this experiment is that almost even hydrogen concentration at the cathode is detected with respect to current densities for all cases. When transforming this data with the flow rate of air supplied, the hydrogen crossover rate is gradually increased as the current density is increased. It means that the mass of the hydrogen gas transferred is increased for higher current densities.

1. Introduction

The polymer electrolyte membrane fuel cell (PEMFC) system has attracted a considerable attention as one of the future power sources. While great efforts have been made for successful commercialization, there still remain unresolved points which need to be improved. Among them, durability issue is the most critical problem [1–6].

In this sense, investigation on the crossover phenomenon, which is strongly related to the durability of the polymer electrolyte membrane fuel cell system, is imperative [7–9]. The crossover occurs inevitably in the system due to the porosity of the membrane. Gas crossover causes a durability problem on materials in the fuel cell [10–14]. Inaba et al. studied the effect of the gas crossover on the durability of the material. They found that the hydrogen crossover makes the performance of the system lowered. On the other hand, the oxygen crossover has much greater effects on deterioration of the membrane than the hydrogen crossover [15]. Kocha et al. analyzed the effect of the nitrogen crossover on the fuel cell system. Through that research, they determined that the local fuel starvation is caused by the nitrogen crossover and it is the reason for the cathode catalyst degradation [16].

With these results from the previous studies, many researches have attempted actively to find the effects of the operating parameters on the crossover rate in fuel cells [12,17–20]. The effect of the temperature and relative humidity was discovered by Inaba et al. [15]. Furthermore, Baik et al. studied the effect of the operating parameters on the hydrogen crossover and suggested the quantitative influence of each parameter to the crossover rate [17]. However, researches about the crossover phenomenon, for the most part, have dealt with the open circuit condition [21–24]. Because

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it seems that the effect of the crossover rate is especially important for open circuit condition, therefore, the regulation guideline released by DOE (Department of Energy, U.S.A.) is also established based on the open circuit condition. However, measuring the crossover rate with different current densities is very imperative because electrical load could change the environment, such as concentration gradient, water production, that affect crossover phenomenon. Furthermore, this is considerably important for not only the durability of the membrane, but also the safety of the system. Thus, departed from the usual aspect of previous research, the limitation for gas crossover rate should be legislated based on the diverse current densities.

For the first time, the hydrogen crossover rates in PEMFCs according to diverse current densities are experimentally investigated in this study. From the previous study, it was revealed that clamping pressure and relative humidity have the biggest influence on crossover among the many operating parameters [17]. Therefore, the hydrogen rates are measured for different clamping pressure and relative humidity conditions. The hydrogen crossover rate with different current densities will be specified by this research, and it gives a substantial measuring technique about the hydrogen crossover rate. The strong relationship between the gas crossover and water contents in the fuel cell will be found through this research.

2. Experimental

2.1. Single fuel cell preparation

In this study, the single PEMFC with active area of 25 cm^2 was used. This single fuel cell consists of a membrane, gas diffusion layers, gaskets, bipolar plates and end plates. The Gore™ PRIMEA® series 57 membrane electrode assembly (MEA) (W.L. Gore & Associates, Elkton, MD, USA), whose membrane is 18 μm thick and 0.2, 0.4 mg/cm^2 Pt loading for anode and cathode, relatively, was used with the Sigracet® 35 BC gas diffusion layers (GDL) (SGL Carbon Weisbaden, Germany) and Teflon® gaskets [25]. The graphite bipolar plates which have the channels of serpentine flow type were used. To prevent gas leakage, the rubber O-rings equipped on end plates were used. Lastly, all prepared parts were uniformly assembled by a torque wrench.

2.2. Experimental apparatus with single fuel cell

The experimental setup for this experiment is shown in Fig. 1. In order to control the temperature, the temperature controllers

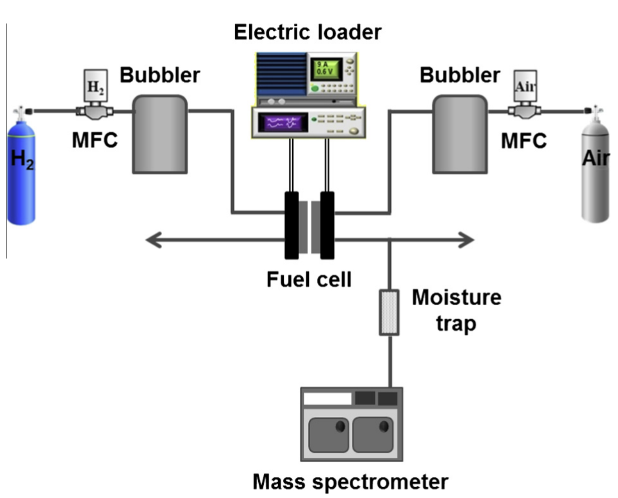

Fig. 1. Schematic diagram of the experimental apparatus with single fuel cell.

Table 1
Experimental conditions.

Operating parameters	Values
Stoichiometric ratio of hydrogen	1.5
Stoichiometric ratio of air	2.0
Temperature (°C)	65
Relative humidity (%)	50/100
Clamping pressure (N m (lbf·in))	7.91 (70)/9.04 (80)
Current density (A/cm^2)	0–1.0

(UT 550, Yokogawa, Japan) were used. The temperature of the single fuel cell was measured by T-type thermocouple (TC), which is inserted in both bipolar plates. Additionally, the same thermocouples were put into both sides of bubble-type humidifiers and gas lines to manage the relative humidity condition. To control the mass flow rate for a fixed stoichiometric ratio, mass flow controllers (HI-TEC MFC, Bronkhorst, The Netherlands) were used, and the pressure was monitored by pressure transducers.

For conducting the activation process and measuring an electrochemical performance, the electric loader (PLZ 1004WA, Kikusui Electrons, Yokohama, Japan) was equipped for the single fuel cell system setup.

To detect the hydrogen crossover rate, the on-line quadrupole mass spectrometer (HPR-20 QIC, Hiden Analytical, Warrington, UK) was used which can analyze and measure the gas concentration by the electron impact ionization via thermionic emission from the hot filament. The flush line for small amount of nitrogen gas was added with a three-way valve for accurate detection.

2.3. Experimental procedure for measuring the hydrogen crossover rate

To begin with, the activation process to make a membrane be at a steady conditioned state was conducted. This process was performed when the temperature and the relative humidity were 65 °C and 100% for both gases and the stoichiometric ratio was 1.5 for hydrogen and 2.0 for air. After this, calibration process was carried out for quantitative measurements.

After all groundwork was done, the port from the mass spectrometer was connected to the cathode outlet to measure the hydrogen crossover rates. At the same time, the performance and the hydrogen crossover rate of the single fuel cell were measured at the condition presented in Table 1. To analyze the crossover rate pattern accurately, detecting time is extended. Then the value for the hydrogen concentration is estimated as the average value from the specific time interval.

2.4. Data analysis

In this experiment, the hydrogen concentration is detected from the cathode outlet. However, the hydrogen flow rate is continuously changed according to the different current density. Therefore, the hydrogen concentration in the unit of ppm needs to be converted into the hydrogen crossover rate in the unit of mol/s cm^2. Conversion of hydrogen concentration into hydrogen crossover rate is plotted in Fig. 2. In this figure, the actual amount of the transferred gas is displayed.

3. Results and discussion

3.1. Hydrogen crossover rate for several clamping pressures

From the previous study, it is known that the clamping pressure has primary effect on the hydrogen crossover rate under an open circuit condition [17]. However, this relation between the