



Local resolved investigation of hydrogen crossover in polymer electrolyte fuel cell



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ABSTRACT

In this study, the effects of temperature, pressure and relative humidity (RH) on hydrogen crossover rate from anode to cathode of a PEMFC is investigated. Segmented cells are used to measure the local hydrogen crossover current density ($j_{\text{H}_2}^{\text{cross}}$) distribution. The results present approximate linear increase of the hydrogen crossover rate with increasing temperature and hydrogen back pressure with rates of $0.038 \text{ mA cm}^{-2} \text{ K}^{-1}$ and $3.33 \text{ mA cm}^{-2} \text{ bar}^{-1}$, respectively. Generally, slightly increased H_2 crossover is observed in gas inlet areas than cell average. Unlike the approximate linear relationship between temperature or pressure with hydrogen crossover, the effect of relative humidification on hydrogen crossover is more complex with different increasing rate before fully humidification and dramatic decline at excessive humidification. It is demonstrated that segmented cells can be advantageously applied to study local H_2 crossover of intact MEAs.

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1. Introduction

Proton Exchange Membrane Fuel Cells (PEMFC) have received considerable attention from researchers in the recent decades as a source of clean energy and as an alternative to conventional internal-combustion engines in automobile and transportation applications due to its high power density, high efficiency, zero emissions, nonmoving parts and silent operation. The main advantage of PEMFC arises from the emission free electricity generation by reacting oxygen and hydrogen to produce water and heat as the only byproducts [1–3]. However, PEMFC still faces several difficulties, such as high cost due to use of expensive materials and limited durability related to component degradation during operation [4]. Thus, a lot of emphasis has been given to study, understand, and alleviate these issues [5–7].

The polymer ion conducting membrane is a key component of membrane electrode assembly (MEA) of a PEMFC. In recent years thinner membranes have been developed to increase ionic conductivity and, hence, to increase the cell performance. However,

with decreased membrane thickness other problems have arisen, such as reactant crossover (especially at low current densities) and reduced mechanical stability of the membrane.

Hydrogen crossover is the diffusion of hydrogen from the anode to the cathode through the membrane which occurs faster than the diffusion of other gases. Several groups [8–11] have investigated the effect of hydrogen crossover. Hydrogen crossover has at least three negative effects on fuel cell operation: fuel efficiency reduction, cathode potential depression, and aggressive peroxide radical formation [12]. Specifically, the hydrogen which crosses over can directly react with oxygen at the cathode surface, resulting in reduced cell voltage due to development of so called mixed cathode potentials [13,14]. H_2 and O_2 could also react directly at the cathode producing peroxide radicals at the same time, which not only attack the catalyst layer but also the membrane, causing significant catalyst layer and membrane degradation [15]. Wang et al. [16] demonstrated via ex-situ NMR (nuclear magnetic resonance) and FTIR (Fourier transform infrared spectroscopic) analysis of Nafion[®] based MEAs that the membrane degradation was originated from the decomposition of polymer main chain. It is speculated that with the increased loss of membrane units, small bubbles with the diameter of several microns started to form inside the membrane itself. These bubbles made the membrane vulnerable to hazards of

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gas crossover, which further led to a catastrophic failure of the proton exchange membrane. There are mainly two ways to form hydrogen peroxide, one being oxygen reduction at the cathode, the other based on the crossover of oxygen from the cathode to the anode. The hydrogen peroxide diffuses into the membrane and reacts with metal ions, present as impurities in the membrane to form HO• or HOO•, which can attack the polymer and degrade the membrane [17,18]. In addition, hydrogen radical species have also been reported can degrade the membrane [19]. Therefore, the measurement of hydrogen crossover is of great importance for the fundamental understanding and practical mitigation of fuel cell degradation and membrane failure.

Effects of operating conditions on the hydrogen crossover has been investigated by several groups [15,20,21] but the detailed mechanism and locally evolving process of hydrogen crossover during the changing of operating conditions have not been clarified. In this study, we applied segmented cell measurements, which are powerful tools for in-operando monitoring of current density distribution [22–26], for locally resolved monitoring of the hydrogen crossover current density distribution. Segmented cells have been already demonstrated to be suitable to detect large amounts of hydrogen crossover due to pinholes in the membrane [24]. To the best of our knowledge, however, they have not yet been used to investigate effects of cell temperature, back pressure and relative humidification on local hydrogen crossover distribution in intact MEAs.

2. Experimental

In this section, the test objects and segmented cell technology are firstly described. Afterwards the electrochemical method of measuring hydrogen crossover and methods of measuring the effects of temperature, back pressure and relative humidity on hydrogen crossover are described separately.

2.1. Test objects

Two different membrane electrode assemblies (MEAs) were used as test objects. MEA1 is a commercially available MEA from Wuhan Xinyuan Corporation with a perfluorosulfonic acid (PFSA) membrane with a thickness of 25 μm . The active electrode area was 50 cm^2 . Single channel serpentine flow fields were applied on both electrodes of the cell. In order to keep the MEA, the gaskets and the segmented board in good contact, clamping pressure of 8 bars was provided by cylinder compression. Relative humidity was controlled by passing reactants through bubbler humidifiers. MEA2 is a commercially available product from Johnson Matthey Fuel Cells. The membrane thickness was 20–25 μm and the active electrode area was 25 cm^2 . The experiments were conducted on a homemade single cell test bench at DLR (see affiliation above) in Stuttgart, Germany, using single channel serpentine flow fields. 50 cm^2 and 25 cm^2 cell plates are used for MEA1 and MEA2, respectively. The test samples were conditioned at 1000 mA cm^{-2} with fully humidified gases for 20 h. Stoichiometry ratios were 1.5 and 2.0 for hydrogen and air, respectively.

2.2. Segmented cell technology

To investigate the locally resolved hydrogen crossover current density, the segmented printed circuit board (PCB) technology [22,24,27] was adopted. The segmented flow field plate integrated with temperature sensors was installed between the MEA and the anode current collector plate. Our previous study [24] ascertained the assumption of identical current density distributions in anode and cathode by integrating segmented bipolar plates on both sides

of the cell. The identical current density distributions obtained indicate negligible lateral currents due to high conductivity perpendicular to the membrane plane compared to the in-plane conductivity and the anode and cathode current density distributions are equivalent. The current density distribution data were collected using a data acquisition unit consisting of a multiplexer and a digital multimeter. In this study, two segmented plates with the same segment configuration but different segment size were used for MEA1 and MEA2, respectively.

2.3. Measuring hydrogen crossover rate

After the conditioning procedure a nitrogen flow was introduced into the cathode to remove the air and the anodic H_2 flow was set at a constant rate. The specific flow of nitrogen was 120 ml min^{-1} for MEA1 and 60 ml min^{-1} for MEA2. The hydrogen flow was 380 ml min^{-1} for MEA1 and 190 ml min^{-1} for MEA2. After 20 min of N_2 flushing, the cathode potential was below 120 mV. Then a Zahner IM6 potentiostat was connected to the fuel cell for H_2 crossover measurements, with the working electrode probe connected to the cathode and the counter/reference electrode probes connected together to the anode. The current produced from the oxidation of crossed H_2 from the anode was measured at a potential of 0.4 V. At this cathode potential, all H_2 that has crossed over from the anode to the cathode should be completely oxidized, producing a current ($j_{\text{H}_2}^{\text{cross}}$) indicating the amount of hydrogen that has crossed over. The molar permeation flux density (ϕ) of hydrogen through the membrane is determined by the measured crossover current densities ($j_{\text{H}_2}^{\text{cross}}$) using Faraday's law

$$\phi = \frac{j_{\text{H}_2}^{\text{cross}}}{zF} \quad (1)$$

Throughout the whole experiment the hydrogen crossover current density distribution was recorded using a segmented plate.

2.4. Effects of temperature and back pressure on H_2 crossover rate

MEA1 was used in this section. Throughout the experiment the RH was kept at 100% for both electrodes. The investigated cell temperature range was from 50 $^\circ\text{C}$ to 80 $^\circ\text{C}$ and the investigated back pressure range was from 1.4 bars to 1.9 bars absolute pressure. Throughout the experiment, $j_{\text{H}_2}^{\text{cross}}$ was recorded by a 50 cm^2 segmented plate.

2.5. Effects of relative humidity on H_2 crossover rate

To analyze the effect of relative humidity on hydrogen crossover MEA2 was used. The RH range, controlled by adjusting the dew point temperature of bubbler humidifiers, was from 50% to excessive humidification. Back pressures were kept at 1.5 absolute bars for both electrodes and cell temperature was kept constant at 80 $^\circ\text{C}$. $j_{\text{H}_2}^{\text{cross}}$ distribution was recorded throughout the experiment using a 25 cm^2 segmented plate.

3. Results and discussion

3.1. Effects of cell temperature on H_2 crossover

Fig. 1 shows the H_2 crossover current density $j_{\text{H}_2}^{\text{cross}}$ in cell temperature range of 50 $^\circ\text{C}$ –80 $^\circ\text{C}$ at 100% relative humidity. In the individual panels $j_{\text{H}_2}^{\text{cross}}$ is plotted as an average value of the entire cell (A), gas inlet area (B), center flow field area (C), and gas outlet area (D). The data is determined from segmented cell measurements depicted in Fig. 2. In accordance with Inaba et al. [28] it was

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