Applied Energy 101 (2013) 560–566

Contents lists available at SciVerse ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy

Local measurements of hydrogen crossover rate in polymer electrolyte membrane fuel cells

Kyung Don Baik [a], Im Mo Kong [a], Bo Ki Hong [b], Sae Hoon Kim [b], Min Soo Kim [a,*]

[a] School of Mechanical and Aerospace Engineering, Seoul National University, Seoul 151-744, Republic of Korea
[b] Fuel Cell Vehicle Team 1, Eco-Technology Center, Hyundai-Kia Motors, Gyeonggi-do 446-912, Republic of Korea

HIGHLIGHTS

▸ Hydrogen crossover appears to occur mostly near the gas inlet region.
▸ The hydrogen crossover rates increase with decreasing the nitrogen flow rates.
▸ Hydrogen crossover increases with the increase in both cell temperature and RH.

ARTICLE INFO

Article history:
Received 2 April 2012
Received in revised form 12 June 2012
Accepted 14 June 2012
Available online 21 July 2012

Keywords:
Hydrogen crossover
Local measurement
Polymer electrolyte membrane fuel cell
Open circuit voltage

ABSTRACT

Hydrogen crossover is the main reason for membrane degradation in polymer electrolyte membrane fuel cells (PEMFCs). In this study, local measurements of the hydrogen crossover rate at the cathode in a PEMFC are investigated to analyze the distribution of hydrogen crossover rates under various temperature and relative humidity (RH) conditions. The bipolar plate for the cathode side is specially designed for local measurements. Results show that hydrogen crossover appears to occur mostly near the gas inlet region, and reduced crossover amounts near the outlet region. The hydrogen crossover rates increase with decreasing the nitrogen flow rates at a given section. The effects of temperature and RH on the hydrogen crossover rate over the entire area of the fuel cell are also analyzed and compared with the results of the open circuit voltage (OCV). The results show that the hydrogen crossover rate increases with the increase in both cell temperature and RH, resulting in a decrease in the OCV.

1. Introduction

Membranes are usually used as an electrolyte in polymer electrolyte membrane fuel cells (PEMFCs). The main functions of membranes are to transfer protons from the anode to the cathode in PEMFCs and form a barrier between the anode and cathode reactants. However, gases inevitably permeate through the membrane barrier, which is referred to as the 'gas crossover' phenomenon. Gas crossover occurs due to concentration and pressure gradients between the anode and cathode sides. The hydrogen concentration in the anode side is usually higher than that in the cathode side; thus hydrogen is transported from the anode to the cathode side by diffusion [1]. Nitrogen and oxygen are also transported from the cathode to the anode side in the same manner. There are several theories to explain the mechanism of gas crossover through the membrane. Yeager et al. [2] suggested a three-region structure model for a Nafion® membrane consisting of the fluorocarbon phase region, interfacial zone and ionic clusters formation region. They reported that gases permeate through Nafion® membranes in the intermediate region. Sakai et al. [3] suggested another theory of gas crossover by using an ionic cluster-network model. They claimed that the gas permeates mainly through the water-containing ion cluster regions of the membrane. However, these two mechanisms are still controversial because of the complexity of the membrane structure, as Kocha et al. [4] mentioned. Thus further research of the gas crossover mechanism is needed.

Gas crossover is an unavoidable phenomenon in PEMFCs, having attracted much interest and research in recent years. Cheng et al. [5] measured the hydrogen crossover rate in high-temperature PEMFCs under various operating conditions by using a steady-state electrochemical method. Vilekar and Datta [6] theoretically analyzed the role of hydrogen crossover in PEMFCs. They mentioned that hydrogen crossover can explain the entire potential loss from the standard-state reversible voltage of 1.23 V under open-circuit conditions. Inaba et al. [7] reported that hydrogen crossover is the main cause of membrane degradation. They suggested the possible degradation mechanisms due to hydrogen crossover: thermal degradation caused by heat generation upon

* Corresponding author. Tel.: +82 2 880 8362; fax: +82 2 883 0179.
E-mail address: minskim@snu.ac.kr (M.S. Kim).

0306-2619/$ - see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.apenergy.2012.06.034

Nomenclature

J	gas permeation rate [mol s^{-1} cm^{-2}]
k	permeability coefficient [mol m^{-1} s^{-1} Pa^{-1}]
l	membrane thickness [μm]
P	partial pressure [Pa]
Superscript	
A	anode side
C	cathode side
Subscript	
H_2	hydrogen gas

direct combustion of hydrogen and oxygen, and chemical degradation by hydrogen peroxide. Bessarabov and Kozak [8] reported that hydrogen crossover lowers fuel cell efficiency and generated the oxygen activation, which degrades the membrane on the cathode side of the fuel cell. Nam et al. [9] numerically investigated the effects of hydrogen and oxygen crossovers through the membrane in PEMFCs. Researches on nitrogen and oxygen crossovers have been also conducted. Baik and Kim [10] measured nitrogen crossover rates experimentally and estimated nitrogen permeability coefficients under various temperature and relative humidity (RH) conditions. Ahluwalia and Wang [11] analyzed the buildup of nitrogen in the anode recirculation system and determined its effect on the performance of the PEMFC stacks. Peron et al. [12] extracted the oxygen permeability data of a solid state electrochemical cell to investigate the properties of newly-developed Nafion® 211 membranes.

Most researches mentioned above only focused on the gas crossover over the entire area of the membrane in PEMFCs, but not over a local area. For example, Cheng et al. [5] calculated the hydrogen permeability coefficient based on the whole area of the membrane. Other researchers [6–9] also considered hydrogen crossover phenomenon over the entire area. However, the hydrogen crossover rate over a local area may be different when the local operating conditions are different. To design highly-durable stacks for fuel cell vehicle applications, the local distribution of the hydrogen crossover rate in fuel cells needs to be understood in depth, because hydrogen crossover can cause local degradations of the membrane. Recently, several local measurement techniques [13–18] have been developed for PEMFCs, but most of them have been applied to measure the local current in the PEMFC rather than the local gas crossover rate. Local measurements of the hydrogen crossover rate can elucidate where membrane degradations occur. Therefore, local measurements of the hydrogen crossover rate need to be studied.

In this study, local measurements of the hydrogen crossover rate in PEMFCs have been conducted to analyze the distribution of the hydrogen crossover rate under various temperature and RH conditions. Furthermore, the effects of operating conditions on hydrogen crossover are investigated.

2. Experimental method

2.1. Single fuel cell and bipolar plate design

A single PEMFC with an active area of 25 cm^2 was used for the local measurements of the hydrogen crossover rate in this study. The graphite bipolar plates with serpentine flow channels were used for both anode and cathode flow fields. As shown in Fig. 1a, the single PEMFC consisted of end plates, graphite bipolar plates, gas diffusion layers (GDLs), gaskets and a membrane electrode assembly (MEA). Bipolar plates and end plates were specially designed for local measurements of the hydrogen crossover rate. A GDL sample with a nominal thickness of 325 μm was obtained from a commercial manufacturer, and consisted of both a micro-porous layer and a macro-porous substrate, which were both wet-proofed by hydrophobic treatment. The macro-porous substrate of the GDL sample was made from carbon fiber papers. Commercially-available perfluorinated sulfonic acid MEA was used in this study. Both the anode and cathode of the MEA were composed of typical Pt/C catalysts, and the Pt loadings of the anode and cathode were both 0.4 mg Pt cm^{-2}. Rubber type O-rings and Teflon® gaskets were also used to prevent gas leakage. A torque wrench, set at 6.78 N m, was used to uniformly compress the single cell assembly. A photo of the single fuel cell used in this study is shown in Fig. 1b. For local measurements of the hydrogen crossover rate in the PEMFC, a bipolar plate was specially designed for the cathode side, as shown in Fig. 2. The anode bipolar plate was composed of conventional five-serpentine flow fields, while the cathode bipolar plate was modified to have parallel and five-serpentine flow fields with gas sampling ports. The cathode flow field had a channel width and depth of 1.0 and 0.9 mm, respectively, and the land width was 1.0 mm. The design parameters of the bipolar plate for

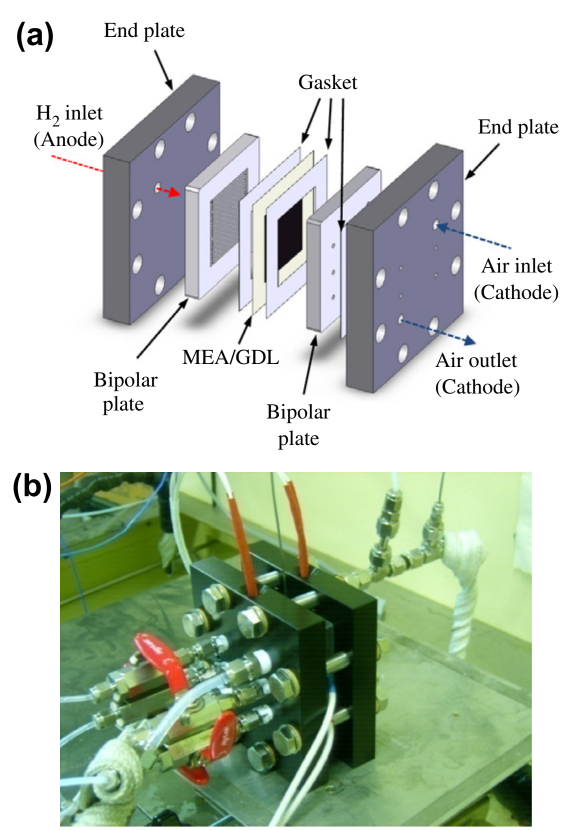

Fig. 1. Schematic representation of the single fuel cell used in this study: (a) an magnified view and (b) a photo of the fuel cell assembly for local measurement of hydrogen crossover.