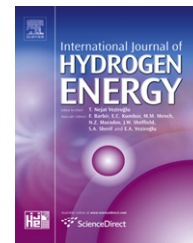




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Development of a galvanostatic analysis technique as an in-situ diagnostic tool for PEMFC single cells and stacks

Kug-Seung Lee^a, Byung-Seok Lee^a, Sung Jong Yoo^a, Soo-Kil Kim^b, Seung Jun Hwang^a, Hyung-Juhn Kim^a, EunAe Cho^a, Dirk Henkensmeier^a, Jeong Woo Yun^c, Suk Woo Nam^a, Tae-Hoon Lim^a, Jong Hyun Jang^{a,*}

^a Fuel Cell Research Center, Korea Institute of Science and Technology (KIST), Seoul 136-791, Republic of Korea

^b School of Integrative Engineering, Chung-Ang University, Seoul 156-756, Republic of Korea

^c School of Applied Chemical Engineering, Chonnam National University, Gwangju 500-757, Republic of Korea

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ABSTRACT

A new galvanostatic analysis technique was developed for PEMFC single cells and stacks, while conventional potentiodynamic techniques, such as cyclic voltammetry for an electrochemical active surface area (EAS) and linear sweep voltammetry for a crossover current (i_{H_2}), cannot be directly utilized for stacks. Using a developed relationship for double-layer charging region, the i_{H_2} and C_{dl} (double-layer capacitance) of a PEMFC single cell could be determined from the galvanostatic data under an atmosphere of nitrogen (cathodes) and hydrogen (anodes). Then, simply from the elapsed time in hydrogen adsorption/desorption region, EAS or roughness factors could be analyzed for a PEMFC single cell. For a 5-cell PEMFC stack, it was experimentally confirmed that the same analysis technique can be applied to analyze performance distribution in PEMFC stacks. As the characteristics of catalyst layers (EAS and C_{dl}) and polymer electrolyte membranes (i_{H_2}) of individual cells can be analyzed without stack disassembly, the developed galvanostatic technique is expected to be utilized for the degradation study and performance monitoring of practical PEMFC stacks.

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1. Introduction

The polymer electrolyte membrane fuel cell (PEMFC) is considered a very promising technology for both stationary and mobile applications owing to its advantages such as high power density, absence of noxious emissions, quiet operation, as well as quick startup and shutdown. Although the technical issues of system cost and long-term durability are delaying its commercial success, PEMFCs are generally expected to be commercialized, in the near future, as a key technical element of the hydrogen economy. For the enhancement of the long-

term durability, a proper diagnosis of practical PEMFC single cells and stacks is required [1,2]. If the degradation behaviors of each cell and each component in PEMFC stacks can be separately analyzed during operation (in-situ), it will be possible to reveal the detailed degradation mechanism of practical PEMFC systems, providing a reasonable and effective strategy to enhance PEMFC durability.

As in-situ diagnosis tools, electrochemical techniques have been widely utilized for PEMFC single cells: polarization curve (I–V curves), current interruption (CI), electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV), and

* Corresponding author. Tel.: +82 2 958 5287; fax: +82 2 958 5199.

E-mail addresses: jonghyun.jang@gmail.com, jhjang@kist.re.kr (J.H. Jang).

linear sweep voltammetry (LSV) [1–7]. For example, the degradation of proton exchange membranes (PEMs) can be monitored by LSV (crossover current), CI (ohmic resistance), and EIS (ohmic resistance), while the degradation in the catalyst layers (CLs) can be characterized by EIS (triple-phase boundary) and CV (Pt/ionomer interfacial area). After completion of the cell operation, further analysis can be carried out by various physical/chemical techniques, including infrared spectroscopy (IR) [8–10], scanning electron microscopy (SEM) [8–11], transmission electron microscopy (TEM) [8,12,13], X-ray diffraction (XRD) [8,13,14], and X-ray photoelectron spectroscopy (XPS) [8,15]. However, for these physical/chemical techniques, sampling from disassembled MEAs is required, and therefore, these methods cannot be utilized for in-situ monitoring of membrane-electrode assembly (MEA) degradation during cell operation.

In the case of PEMFC stacks, galvanostatic techniques are preferred as cells are serially connected in most systems. Therefore, the stack evaluation is usually made by measuring the stack voltage and cell voltage distribution under galvanostatic control, as well as by EIS measurement with current perturbation. Potentiodynamic techniques that are used for single cells, such as CV and LSV, cannot be directly applied in the analysis of PEMFC stacks, because the voltage distribution in the stack is generally not uniform and has different cell performances and degradations. For Pt/C catalysts in a half-cell configuration, it has been reported that the galvanostatic measurement can be utilized to collect the differential capacity, which is equivalent to CV data, and to calculate the electrochemical active surface area (EAS) [16]. However, the use of this galvanostatic analysis was not validated for the PEMFC single cells and stacks, where the effect of hydrogen crossover through PEMs and multiple cells with different performances should be considered in the analysis of galvanostatic data.

In the present study, a galvanostatic analysis technique under an atmosphere of nitrogen (cathodes) and hydrogen (anodes) was developed as an in-situ diagnostic tool for PEMFC single cells and stacks. For the direct conversion of galvanostatic data to differential capacity [16], the effect of hydrogen crossover was discussed to verify whether such EAS calculation is applicable to the practical PEMFC MEAs. Then, a new analysis procedure for galvanostatic data was developed to calculate important parameters of MEA characteristics, including the EAS and crossover current. The developed galvanostatic analysis technique was validated by the analysis of a 5-cell PEMFC stack, where the distributions of EAS, crossover current, and double-layer capacitance values could be obtained for each cell in the stack.

2. Experimental

2.1. Electrochemical measurements of a single cell

Using an NRE-212 membrane (DuPont Inc.), an MEA was prepared by the decal transfer method as described in a previous paper [17]. The Pt loadings were 0.157 mg/cm² and 0.144 mg/cm² for the cathode and anode sides, respectively, with an active area of 25 cm². The Nafion ionomer content

(DuPont Inc., EW 1100, 21 wt.%) was controlled to be 30 wt.% based on the total solid content.

For electrochemical measurements, a single cell was assembled by using the prepared MEA, gas diffusion layer (SGL carbon Inc., Sigracet 10BC), Teflon gaskets, and graphite blocks [17,20–22]. After installation at a fuel cell test station (WonAtech, Smart II), the single cell was activated by potential cycling at three voltages of open circuit voltage (OCV), 0.6 V, and 0.4 V for 6 h. Under fully humidified hydrogen (300 ml/min, anode) and nitrogen (120 ml/min, cathode), CV was performed between 0.08 and 1.2 V at scan rates of 20, 50, 100, and 200 mV/s (Science Instruments, BioLogic HCP-803). Then, galvanostatic measurements were performed at constant currents of 50, 100, 200, and 300 mA (2, 4, 8, and 12 mA/cm² of current densities) with initial and cutoff voltages of 0.08 V and 1.2 V, respectively. The single cell temperature was maintained at 65 °C.

2.2. Electrochemical measurements of a 5-cell stack

A 5-cell stack was fabricated using commercial MEAs with an active electrode area of 25 cm², commercial gas diffusion layers (Sigracet 10BC), and home-made graphite bipolar plates with serpentine flow patterns. Prior to the analysis, the assembled 5-cell stack was discontinuously operated with current variations between 0 and 20 A. After the 5-cell stack was activated at 2 V for 5 h, corresponding to an average cell voltage of 0.4 V, fully humidified hydrogen (300 ml/min) and nitrogen (120 ml/min) were supplied to the anodes and cathodes, respectively, for the galvanostatic analysis. With initial and cutoff voltages of 0.08 V and 1.2 V, respectively, the voltage variations of individual cells at a constant current of 200 mA (current density of 8 mA/cm²) were recorded one at a time. Similar data collection was repeated at higher currents of 300 and 400 mA. The stack temperature was maintained at 65 °C.

3. Results and discussion

3.1. CV analysis of PEMFC single cells

In a standard CV measurement, the electrode potential is swept back and forth at a fixed scan rate between two voltage limits (V_c and V_a), as shown in Fig. 1a, and the corresponding currents are recorded. Here, $t_{1/2}$ and t_1 represent the half-cycle time and full-cycle time, respectively. For the CV measurement of a PEMFC single cell, the working electrode (usually the cathode) is flushed with inert gas (nitrogen or argon), while the other electrode (usually the anode) is flushed with hydrogen, making it act as a dynamic hydrogen electrode (DHE).

Fig. 2a shows cyclic voltammograms at various sweep rates for the cathode of a PEMFC single cell (nitrogen in cathode; hydrogen in anode). Here, two redox peaks are noticeable, which represent the electrochemical adsorption/desorption of hydrogen on platinum (around 0.2 V) and Pt surface oxidation/reduction (>0.8 V for an anodic sweep and >0.6 V for a cathodic sweep). The electrochemical hydrogen adsorption/desorption is observed in a voltage range between 0 and 0.4 V,

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