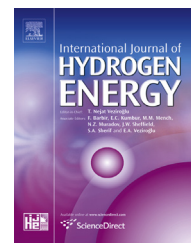Available online at www.sciencedirect.com

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Analysis of the spatially distributed performance degradation of a polymer electrolyte membrane fuel cell stack

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ABSTRACT

Herein we report the spatially uneven degradation of a polymer electrolyte membrane fuel cell (PEMFC) stack operated under load variation. Fifteen sub-membrane electrode assemblies (sub-MEAs) at various cell positions and various points within each cell were obtained from the original MEAs employed in the fuel cell stack. Polarization curves and the voltammetric charge of these MEAs were measured in order to correlate localized performances with the redistributed electrochemically active surface on Pt using the polarization technique and cyclic voltammetry. Several *ex situ* characterizations including electron probe microanalysis, environmental scanning electron microscopy, and X-ray diffraction were also performed to find evidence, supporting the inhomogeneous degradation of the fuel cell stack. Possible routes and processes for the non-uniform stack degradation during the PEMFC stack operation will also be discussed.

Introduction

As the scale and scope of industrial and economic activities continue to increase, the depletion of natural resources and environmental pollution are becoming serious issues. Many investigations are being actively conducted to improve the efficiency of alternative energies that are regarded as "clean energy". As a potential alternative energy source, hydrogen is generally regarded as an environmentally and climatically clean form of energy [1]. As research towards hydrogen energy increases, the development of fuel cell technology also attracts much attention.

Fuel cells are electrochemical devices that directly convert chemical energy into electricity, and typically use hydrogen as a fuel source [2]. Among the various types of fuel cells,

polymer electrolyte membrane fuel cells (PEMFCs) have received much attention as a promising alternative power source for automobiles, because of its significant advantages including a high efficiency, low operating temperature, fast startup, and zero greenhouse gas emissions. However, with respect to the durability and reliability of PEMFCs, several issues such as membrane failure [3], carbon corrosion [4], Pt degradation [4−6], and impurity adsorption on Pt [7] need to be addressed in order for these devices to be viable for commercialization [3−6,8−18]. Additionally, the non-uniformity of each component due to variable operating conditions such as an uneven distribution of reactants, inappropriate thermal management, and rapid changes in the load may decrease the overall performance and accelerate the degradation rate of an on-board PEMFC stack. For example, Lee et al. investigated flooding in a single cell and stack by visualizing the two-phase flow within the flow channels using three transparent fuel cell fixtures. They claimed that the individual cell performance varied within the stack because of the inhomogeneous distribution of the fuel and water, produced and condensed in the stack [19]. Perez et al. presented a simulation of single-cell performance under a fuel starvation, demonstrating that a sub-stoichiometric amount of hydrogen led to a higher interfacial potential near the fuel outlet [20].

Much effort has been dedicated to analyzing the non-uniformity along the gas flow channel in single cell tests [21−24]. Hwnag et al. mapped the local current distribution of a PEMFC containing segmented current collectors to observe the local current density throughout each segment and subsequently examined the effects of the flow channel pattern, stoichiometry, and relative humidity of the oxidants at the cathode on the current distribution [21]. Similarly, Reshetenk et al. introduced a segmented cell system and investigated the dependency of the potential (or current) distribution throughout the cell on the flow channel parameters and operating conditions [22−24]. Stumper et al. attempted to determine the localized current and electrochemical activity of a single cell by placing several isolated sub-cells at tentatively chosen locations within the membrane electrode assembly (MEA) [25]. In order to analyze the current distribution of a cell as well as between the cells of a PEMFC stack, Pei et al. employed a PEMFC stack composed of 100 MEAs (with a geometric area of 274 cm^2) that was operated under an accelerated stress test for 500 h. Their transmission electron microscopy (TEM) analyses and wettability measurements demonstrated that the local current density of the cell decreased along the flow channel [26].

As mentioned above, most previous investigations have focused on original MEAs rather than fragments of the MEAs to segmentally characterize non-uniformity. In this report, a PEMFC stack was operated with load variation, and three large MEAs were randomly selected from the front, middle, and back portions of the stack. Several sub-MEAs were then snipped off along the flow pattern. In order to understand how the spatially heterogeneous degradation of the PEMFC through the manifold and along the flow channel occurs during the PEMFC stack operation with load variation, all cropped MEAs were tested within a single cell fixture and characterized using various electrochemical and physico-chemical methods.

Experimental

Sample selection

A PEMFC stack containing a series of commercially available MEAs was operated with load variation. As shown in Fig. 1(a), three MEA samples along the manifold were collected and subjected to various characterizations. Five sub-MEAs were then cut from each original MEA and numbered with respect to the flow pattern, as shown in Fig. 1(b).

Single cell operation

Single cells were assembled using Au-plated current collectors, gaskets, and a graphite flow field with an effective area of 9 cm^2. The single cell was then activated at 0.45 V for 20 h. During the activation step, pure hydrogen and air were fed into the anode and the cathode under a constant stoichiometry of 1.5 and 2, respectively. The cell was operated at 65 °C under a steady supply of fully humidified hydrogen and air.

Electrochemical analysis

In order to evaluate the performance degradation, polarization curves of the MEAs were obtained using an automated PEMFC test station (CNL Energy Inc.) and a DC electronic load (ESL-300Z, E.L.P tek). Cyclic voltammetry (CV) experiments were performed using an electrochemical interface (SI1287, Solartron) and an impedance/gain-phase analyzer (SI1260, Solartron). CV measurements under hydrogen at the anode and nitrogen at the cathode were carried out by applying a potential in the range of 0.05−1.2 V with a scan rate of 50 mV s^{-1}. The data obtained from the CV
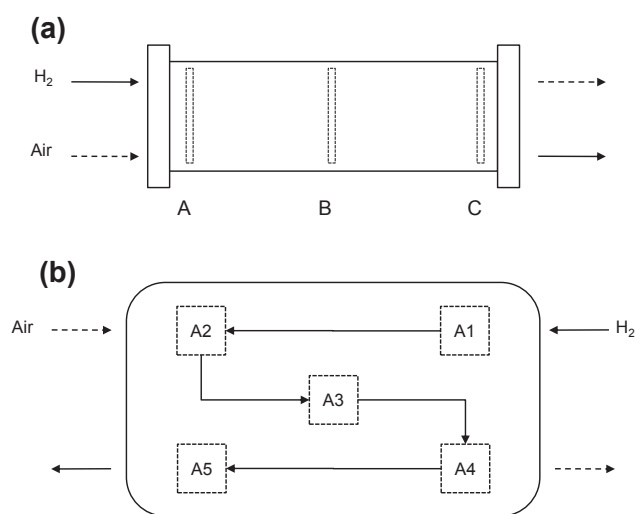

Fig. 1 − (a) Positions of the membrane electrode assembly (MEA) samples within a PEMFC stack, and (b) positions of the sub-MEA samples taken from the MEA.