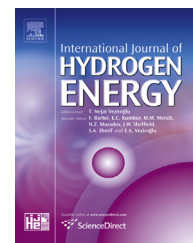




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Effects of carbon corrosion on mass transfer losses in proton exchange membrane fuel cells

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ABSTRACT

Carbon corrosion in proton exchange membrane fuel cells (PEMFC) causes not only kinetic degradation, but also damages to electrode microstructure and hydrophobicity, which can lead to increases in mass transport resistance. While much attention has been paid to catalyst degradation and kinetic losses, the increases in mass transfer loss is also a very serious problem and thus it is the focus of this work. To induce carbon corrosion, accelerated stress test (AST) by holding the cell potential at 1.4 V is used. The AST procedure is interrupted periodically to record cell performance after each period of AST at three different current densities. Experiment results show that at low current density, the decrease in cell voltage is linear with time of AST, but the rate in cell voltage decrease accelerates after some period of AST at medium and high current densities. It is hypothesized that such an accelerated voltage decrease is from the increase in mass transfer loss due to water flooding in the MEA. Further experiments with either reduced inlet air humidification or reduce air flow rate confirm that water flooding in the MEA is the cause for the sharp decline in cell voltages. A phenomenon that the cell voltage increases, or the rate in cell voltage reduction decreases as cell degradation progresses is repeatedly observed. It is determined that such a phenomenon is mainly caused by the enhanced phase-change-induced flow (PCI) due to the higher heat generation rate in a more severely degraded cell. The experimental results also provide some insights on how to optimize operating conditions for degraded fuel cells.

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Introduction

Carbon corrosion in a proton exchange membrane fuel cell (PEMFC) has been recognized as a serious cause in reducing

fuel cell durability and much effort has been devoted to the understanding the mechanisms of carbon corrosion and its detrimental consequences [1,2]. The mechanisms for reverse-current have been particularly clarified, and it is known that reverse current may occur during frequent start-ups and shut-

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downs, reactant starvations, reactant mal-distribution or improper water management [3,4]. Extensive studies have been conducted on the resulting catalyst degradation and kinetic losses [5,6], the interactions between carbon support and platinum catalyst [7,8], hydrophobicity loss of carbon surface [9–11], porous microstructure and morphology damages in the catalyst layer (CL) [12–14] and the interface between the micro-porous layer (MPL) and the gas diffusion layer (GDL) [15].

Corrosion of carbon support in CL causes platinum nanoparticle detachment, migration, electronic isolation or agglomeration, leading to severe reduction in electrochemical area (ECA) [6,16]. Besides, consumption of carbon support is also reported to decrease the thickness of CL [5,7] and even to result in microstructure collapse in the CL [7,13]. Hydrophilic oxide functional groups formed on carbon surface make the carbon surface less hydrophobic [10,11] and alter the morphology and interfaces in CL [17,18]. Carbon corrosion was also reported to occur in GDL and MPL [15,19]. Chen et al. [20] studied the carbon corrosion in GDL with an *ex situ* experiment and found severe structure changes and hydrophobicity loss. Ha et al. [15] conducted an *in situ* experiment to study carbon corrosion in GDL with MPL. The penetration part of MPL into GDL pores was found to be the most vulnerable. Emergence of empty space or gap at the MPL penetration part was observed due to carbon corrosion.

Due to the significant damages of carbon corrosion on electrode porous microstructure, alternation of material physicochemical properties and composition in CL and GDL/MPL, water transport in pores and further mass transfer loss must be affected. It is known that water transport plays a vital role in determining fuel cell performance. Water balance in PEMFC should ensure a good trade-off between flooding avoidance and membrane hydration to reduce both ohmic and mass transfer losses [21]. Hence studies of water transport and water management both in CL and GDL/MPL always draw special attentions. The mechanisms for water transport in multi-scale porous layer, especially the influence of material properties, compositions, porosity, thickness, fabrication method and operating parameters on water transport both in CL and GDL/MPL have been studied extensively [22,23]. The porous structure [24,25], the hydrophobicity [26,27] and interfaces between components [28,29], which are confirmed to be key factors affecting water transport, are all susceptible to carbon corrosion. However, the relationships between carbon corrosion and water transport and further with mass transfer losses in PEMFC are still unclear. Liu et al. [14] measured the limiting current density in a PEMFC with local hydrogen starvation, and sharp current density drop was observed at the starvation area where the carbon corrosion was severe. Park et al. [18] found that the mass transfer limitation increased due to carbon corrosion. Fairweather et al. [30] studied the effect of carbon corrosion on through-plane water transport under different conditions. Pore structure deterioration, drying out effect of the heat generation in corroded cell, and hydrophilic property of the oxidized carbon surface, each differently influences through-plane water transport. Wood et al. [31] proposed a hybridized method to separate the mass transfer over-potential in the cathode GDL and CL during a long term PEMFC operation. However, so far

the change of mass transfer losses caused by carbon corrosion remained inconclusive. The variation of the water transport in porous layers, including CL and GDL/MPL, introduces great uncertainties to water management and control strategy for PEMFCs [30,32].

The objective of present work is to study the effects of carbon corrosion on mass transfer losses in PEMFC. The mechanisms of carbon corrosion on mass transfer losses are further studied by using either lower air humidification or lower air flow rate. The mechanisms are further studied through the hysteresis phenomenon observed during the voltage sweeping procedures.

Experimental

Experimental setup

A single PEMFC with an active membrane electrode assembly (MEA) area of 16 cm² is used in the experimental studies. Nafion[®]212 based catalyst coated membrane (CCM) (Shanghai Pearl Hydrogen Power Source, China) with a catalyst loading of 0.4 mg cm⁻² are used. Two identical pieces of carbon paper (Toray 060, Toray Industries, Inc., Japan) with micro porous layer (MPL) are used at the anode and cathode. Graphite collector plates with machined serpentine flow fields at both sides (co-flow) are used. The width of gas channel and rib is 2 mm and the depth of gas channel is 1 mm. Compression of the single cell is implemented with fuel cell hardware (Fuel Cell Technologies Inc., USA) with heaters and thermocouples to control operating temperature. Fig. 1 is a schematic of the experimental fuel cell.

The experimental system consists of the single fuel cell, a fuel cell test station (FCTS-16, Fuel Cell Technologies, Inc., USA), a Potentiostat/Galvanostat (HCP803, Bio-Logic, Inc., France). Operating parameters, such as cell temperature, reactant gas flow rates, gas humidification and back pressure, can be precisely controlled. The Potentiostat/Galvanostat is used to control cell voltage and current, and conduct electrochemical characterization, including polarization curves and electrochemical impedance spectroscopy (EIS).

Performance characterization

Prior to each set of experiment, the cell is preconditioned at 343 K with fully humidified hydrogen and air at 168 sccm (corresponding to stoichiometry of 3 at current density of 500 mA cm⁻²) and 560 sccm (corresponding to stoichiometry of 4 at current density of 500 mA cm⁻²), respectively. The cell voltage is decreased every 5 min at 0.1 V/step between OCV and 0.2 V until a stable performance has been reached. After the initiation procedure, the cell is considered to be at the beginning of life (BOL) and cell voltages are recorded at three different controlling current densities, i.e. 150 mA cm⁻², 300 mA cm⁻², and 500 mA cm⁻². After the BOL characterization, an AST procedure is conducted. A relative high potential, 1.4 V in this study, is applied to the cell and held for 3 or 6 h. During the AST procedure, anode is supplied with 100% humidified hydrogen and a flow rate of 200 sccm and the cathode is supplied with 100% humidified nitrogen and a flow rate of

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