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Investigation of the recoverable degradation of PEM fuel cell operated under drive cycle and different humidities

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ABSTRACT

Recoverable degradation of a proton exchange membrane fuel cell (PEMFC) under different relative humidities (RHs) after a whole night rest was investigated. A single cell was operated under drive cycle to simulate the working conditions of fuel cell vehicle. It was found that the cell performance decreased after 5 h operation and recovered mostly after one night rest at higher humidities, i.e. 100%, 75% and 50% RH for both cathode and anode sides; while continuous decrease took place at lower humidity, 35%RH. Polarization curve, electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV) and linear sweep voltammetry (LSV) were conducted before and after every 5 h drive cycle for investigating the mechanism of the recoverable degradation. It was found that water content, current density and thermal management might be the main contributions to the performance degradation, by impacting the membrane conductivity, internal resistance, electrode kinetics, and catalyst utilization. A good understanding of voltage recovery phenomenon after several hours rest and its effect on durability will be helpful in improving the reliability and durability of PEMFC.

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Introduction

Proton exchange membrane fuel cell (PEMFC) has the advantages of high power density, low operating temperature, quick startup and low emission, making it applicable in transportation as well as stationary fields [1]. However, nowadays high cost, lack of hydrogen refueling facilities and insufficient

durability are the main obstacles hindering the commercialization of PEMFC. For vehicular PEMFC, its durability has been investigated from various perspectives, such as: long time constant discharging [2,3], load cycling [4–6], specific operational conditions (such as humidity cycling, higher temperature, open circuit voltage (OCV) operation, etc.) [7,8], novel material and stack fabrication approaches (for instance, Pt-perfluorosulfonic acid (PFSA) composite membrane, different

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thickness membrane at the same stack) [9,10]. In the literature, almost all the articles about PEMFC durability focus on irrecoverable degradation attributing to the decay of key materials, like carbon corrosion, catalyst dissolution and aggregation and membrane degradation. Compared to the well explained and understood irrecoverable degradation, recoverable degradation caused by the interruption of continuous electrochemical testing or non-ideal inner water management is also important for the improvement of reliability and durability of PEMFC. However, to our knowledge, limited literature on this issue have been published.

When PEMFC durability experiment is interrupted due to certain reasons, such as electrochemical “in-situ” testing (polarization curve, EIS, CV), equipment maintenance, etc, a phenomenon of recoverable degradation will occur, which has been found in literature [11–16]. The main reason for this phenomenon can be attributed to water and thermal management, which may change the water content in proton exchange membrane (PEM), catalyst layer (CL), and gas diffusion layer (GDL), etc. Water in cell comes either from humidification or electrochemical reaction. If cell is properly hydrated, i.e. in a water balanced state, the conductivity of PEM, as well as the electrochemical performance of CL could be greatly exhibited. On the other hand, dehydration of PEM and flooding in CL would lead to obvious degradation. Recently several studies aiming at the influence of RH on PEMFC performance have been conducted [17–21], however only very limited analyses have been found focusing on the recoverable degradation affected by rest during the interruption of in-situ measurements [22,23]. The mechanism of recoverable influence needs to be better understood.

The purpose of this work is to study the recoverable degradation of fuel cell after overnight rest under different gas humidities which is the main operational factor affecting the water and thermal management compared to temperature, pressure and other factors and find the basic mechanism by “in-situ” testing. Herein drive cycle is used to strengthen the performance degradation, meanwhile makes the recoverable phenomenon detectable.

Experimental

Fuel cell materials and testing apparatus

A single cell with an active area of 50 cm² was used. The membrane electrode assembly (MEA) of the cell included an N211 (DuPont) PEM with catalyst loadings of 0.1 mg_{pt}/cm² for anode and 0.3 mg_{pt}/cm² for cathode; GDLs (TORAY 090) were hot-pressed on both sides. The cell employed a single serpentine channel as the anode flow field and quadruple parallel serpentine channels as the cathode flow field. Gaskets with a thickness of 0.55 mm were used on both sides. The MEA was fully activated prior to all the experiments. An automatic test station (GreenLight G20) was adopted for testing, by which all the operating parameters, including temperature, pressure, humidity, and gas flow rate, could be accurately controlled.

The working temperature of the cell was maintained at 80 °C by controlling the coolant temperature. The inlet

temperatures of hydrogen and air were further guaranteed by tape heaters, meanwhile different humidities (100%, 75%, 50%, and 35% for both anode and cathode sides) were set by adjusting the dew point temperatures of the gases. In all tests, the stoichiometric ratios and inlet gas pressures of anode/cathode were kept to 1.2/2.5 and 60/60 kPag, respectively.

Description of the drive cycle

A drive cycle with a period of 1200 s was designed according to the single cell's response (current density) according to the standard drive cycle, J1015. This drive cycle includes start/stop, idle, part power, rated power and overload operation, and is clearly expressed in Fig. 1. The current density under certain power is used as the control parameter for the fuel cell and a full throttle power, i.e.120% of the rated power is imposed. The capacity of overload (120% rated power) was set at the current density of 1200 mA/cm² and the proportion of OCV operation accounted for 37.91% of the time.

In situ electrochemical measurements

Before and after the drive cycle testing, four “in-situ” electrochemical characterization approaches were conducted to investigate the performance of single cell: polarization curve, electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV) and linear sweep voltammetry (LSV) technologies. The EIS for cell impedance was recorded at 100 mA/cm² galvanostatically within a frequency range 100 mHz–10 kHz and the amplitude was 500 mA. ECSA of the electrodes and hydrogen crossover were calculated respectively by CV and LSV. All the electrochemical results were obtained with a multi-channel potentiostat (VMP2/Z, PAR); meanwhile the cathode was set as working electrode in which pure N₂ (99.99%) was flushed at a flow rate of 0.1 L/min. The scanning range of CV is 0.05–1.1 V with a scanning rate of 20 mV/s. For LSV, the scanning range is 0–0.7 V with a scanning rate of 2 mV/s. After every 5-h durability testing and electrochemical measurements, an overnight rest was performed before studying the recoverable degradation phenomenon.

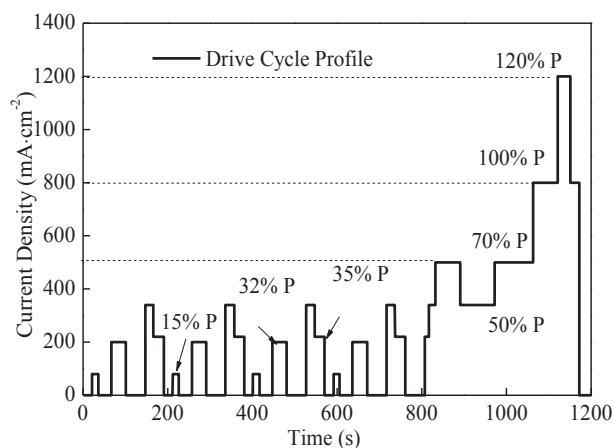


Fig. 1 – Simulated automobile drive cycle profile (period 1200 s).

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