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Impact of dummy load shut-down strategy on performance and durability of proton exchange membrane fuel cell stack

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HIGHLIGHTS

- Segmented cell technology shows the whole performance degradation process.
- Dummy load mitigates performance degradation and enhances durability of PEMFC stack.
- SU/SD process has an obvious impact on single cell, but not on uniformity of stack.

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ABSTRACT

In this paper, the performance and durability of a self-designed proton exchange membrane fuel cell (PEMFC) stack with 5 single cells are investigated with in-situ segmented cell technology. Effect of dummy load application during shut-down process on performance degradation is analyzed through measuring polarization curves and electrochemical impedance spectroscopy (EIS) after different start-up and shut-down (SU/SD) cycles. Scanning electron microscope (SEM) images of membrane electrode assembly (MEA) are taken after the SU/SD cycles. According to the analysis results, it is confirmed that dummy load can take benefit to the durability of PEMFC during the SU/SD process and also the uniformity of current density in different regions for each single cell. However, dummy load may not affect the uniformity of single cells for the stack. This paper contributes to the strategies for promoting performance and durability of PEMFC stack.

1. Introduction

The demand of fossil fuel in the transportation industry is becoming surging. The environment pollution brought by the combustion engine has become a big problem. Governments and research institutes have been exploring new energy vehicles for decades. Fuel cells have attracted a vast amount of attention for their long-life spans, low operation temperature and environment friendly [1–4]. Proton exchange membrane fuel cell (PEMFC) is commonly envisioned to be a promising portable power device and part of transportation power system for its high efficiency and low emission. However, durability, stability and cost which prevent PEMFC from commercial application still need to be solved. Some inevitable operating conditions for automobile application such as dynamic loading, repetitive SU/SD cycles and idling have negative effect on durability of PEMFCs [5]. These adverse operating conditions cause corrosion and chemical reaction of cell components, which deteriorate the electrode material during long term operation [6,7]. Carbon black is widely used as catalyst support in electrodes for its low cost, high surface area and good chemical stability. However, carbon has a low equilibrium potential of carbon corrosion, and the following reactions happen at high electrode potential:

$$\mathrm{C} + 2\mathrm{H_2O} \rightarrow \mathrm{CO_2} + 4\mathrm{H^+} + 4\mathrm{e^-},\ E^0 = 0.207\ \mathrm{V\ vs.\ SHE}$$

$$\mathrm{C} + \mathrm{H_2O} \rightarrow \mathrm{CO} + 2\mathrm{H^+} + 2\mathrm{e^-},\ E^0 = 0.518\ \mathrm{V\ vs.\ SHE}$$

During normal operation the rate of carbon corrosion is negligible, but carbon corrosion can happen at potentials higher than open circuit voltage (OCV) [8,9].

The mechanism of performance degradation during SU/SD process is widely studied and several methods have been proposed to mitigate the performance decay. The most commonly approved mechanism is the 'reverse current decay mechanism' presented by Reiser et al. of United Technologies Corporation (UTC) in 2005 [10,11]. Hydrogen/air interface forms up during shut-down process due to air diffusion and permeation. After shut-down process, anode and cathode flow fields of

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fuel cell are filled with residual hydrogen and air, respectively, then air can diffuse from cathode to anode under the driving force of concentration gradient. Due to the hydrogen/air interface, oxidation and reduction reaction occurs simultaneously at anode and cathode, which yields a high potential up to 1.44 V causing a reverse current and carbon support corrosion. According to the experimental results from Kim et al. [12], cathode potential above 1.0 V will cause oxidation of carbon support and increase of CO_2 amount. At potential above 1.2 V, CO and SO_2 can be detected, which means Pt catalyst is poisoned.

Taniguchi et al. [13] studied the degradation caused by reverse current during air starvation and analyzed the membrane-electrode assemblies (MEAs) through transmission electron microscopy (TEM) analysis and energy dispersive X-ray (EDX) analysis, an obvious decrease of cell performance was observed in a sample degraded by cell reversal for 120 min.

Eom et al. [14] and Dillet et al. [15] studied the carbon corrosion phenomenon in MEAs with different Pt loading and surface area of carbon support during SU/SD process. Pt loading had much more obvious effect on reverse current forming in comparison to operating performance of fuel cell, a lower value of Pt loading at anode and a higher value at cathode significantly diminished the reverse current during the SU/SD process. Besides, a higher rate of carbon corrosion was found in the exit of fuel cell using MEA with bigger surface area carbon support. Operating parameters also had effects on the corrosion of carbon support during SU/SD process. The internal current decreased with the increase of reactant flow rate due to a shorter hydrogen/air interface residence time. A lower relative humidification also could decrease the corrosion rate of carbon support [16].

To mitigate the performance degradation during SU/SD process, more stable catalyst supports have been developed in substitute to carbon black. Carbon materials such as carbon nano tube (CNT) and nano fiber have better chemical stability than carbon black, but the commercial application is still a problem due to the high cost and complicated manufacturing process [13,17,18].

Besides, optimized SU/SD strategies were also developed to reduce the negative impact of hydrogen/air interface. Purging and applying dummy load to consume the residual hydrogen at anode [19,20] and oxygen at cathode [21,22] were two most commonly used strategies to protect carbon support from hydrogen/air interface. Purging strategies include nitrogen or air purging at anode [5], anode exit gas recycling purge and hydrogen purging at cathode. Yu et al. [23] investigated the corrosion of carbon support at anode due to the high potentials during SU/SD process and summarized mitigation strategies against degradation during SU/SD process: reducing the time that hydrogen/air interface exists, such as fuel purging; controlling the potentials below corrosion potential, such as applying an external auxiliary load or an internal short. Qiang shen et al. [24] studied different purging strategies and application of dummy load. They found that purging nitrogen into anode and using dummy load during the shut-down process could mitigate the negative impact of hydrogen/air interface and prevent the formation of high potential at cathode, but purging nitrogen costed long time to remove the residual hydrogen because the desorption of hydrogen was relative difficult. Jithesh P et al. [25] studied the effect of dummy load application on degradation of fuel cell performance and found that shut-down process with dummy load costed half minute in comparison of half hour without dummy load. Yu et al. [26] introduced dummy load to open-ended and closed-ended cells to investigate the effect of cathode exit conditions on the degradation of fuel cell. The result revealed an improvement on durability and lower performance decay in closed fuel cell, the closed cell significantly reduced the decrease in electrochemically active surface area (ECSA). Lin et al. [27] did a better job of controlling the SD time and mitigating degradation by using a two-phase dummy load composed of a linearly declined main load and a fixed small auxiliary load on PEMFC.

Although many relevant researches have been made, studies about strategies applied on stack are limited. Most studies are concentrated on the test parameters [28,29], procedure on the performance of stack [30] and transient response of stack in start-up procedure [31]. Tolarz et al. [32] gave out a mitigation strategy against PEMFC degradation during shut-down procedure, which consisted of isolation of hydrogen with opening anode to external atmosphere, stopping air flow through cathode and large electrical load. Four SU/SD durability tests on stack with 10 cells were conducted and the result showed that only the shut-down strategies with these three elements could effectively reduce corrosive degradation after the PEMFC stopped. Kannan et al. [33] conducted a long term SU/SD experiment on high temperature PEMFCs with 5 single cells. More than 1500 SU/SD cycles were performed to study the degradation effect on repeated test cycles with protective strategy, which could avoid the formation of high potential.

This paper studied the protective strategy of PEMFC stack by conducting two groups of experiments, with or without dummy load in the shut-down process. After 250 cycles in the unprotected group and 1000 cycles in the protected group, the protected group showed much slower decay of performance, which meant better durability. With polarization curves, EIS, SEM and in-situ segmented cell technology, the performance decay of PEMFC was confirmed during SU/SD process. And impact of SU/SD process on single cells of the stack was discussed from different perspectives in this paper.

2. Experimental

2.1. Test bench and PEMFC stack

In this paper, a G60 fuel cells test bench from Canada Greenlight company was used to evaluate the stacks. With HyWARE II software application, the test station provided user interface and controlled the operating parameters such as temperature, pressure, flow rate of reactants, electronic load and so on. Besides, the test bench could also run the scripts designed by users and change the operating condition accordingly.

Structure of the experimental stacks is shown in Fig. 1, the same as the reported one from Lin's group [34]. The bipolar plates (1–5) made from stainless steel were 4 mm thick, and the flow field and coolant gallery were milled on them. Stainless steel was chosen for its excellent electrical conductivity and corrosion resistance. Serpentine flow field with four channels was used in cathode and serpentine flow field with three channels in anode. Through the main pipes, hydrogen and air would arrive at anode and cathode of each cell, respectively. The anode flow fields of No. 1, No. 3 and No. 5 single cells were carved on the printed circuit boards (PCBs) (A-C). Based on the segmented cell technology, the segmented current density could be measured [35]. The detail of this technology was reported before [36]. Five single cells were assembled together by eight 8 mm screw bolts, the clamping force was gradually added until there was no obvious change of the High

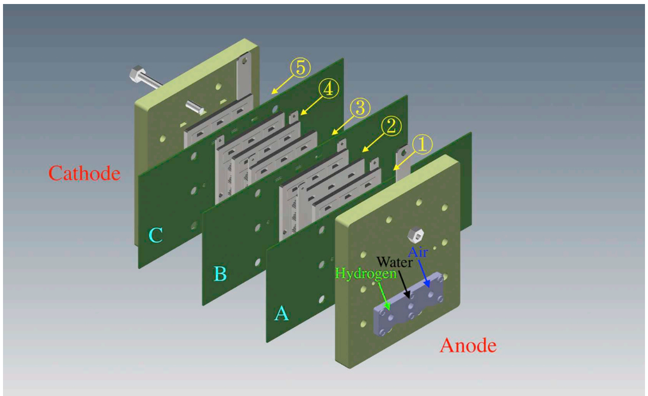

Fig. 1. The structure of self-designed PEMFC stack with in-situ segmented cell technology (A–C: PCBs; 1–5: positions for MEAs).