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Short communication

Analysis of the behavior and degradation in proton exchange membrane fuel cells with a dead-ended anode

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HIGHLIGHTS

- Local current firstly degrades at anode outlet region, then proceeds to the inlet.
- The fuel starvation leads to a high cathode potential and carbon corrosion.
- SEM images reveal thickness reduction and porous structure collapse.
- The dominant factor, leading to the performance decay, is the nitrogen crossover.

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ABSTRACT

Proton exchange membrane fuel cells (PEMFCs) with a dead-ended anode (DEA) can obtain high hydrogen utilization by a comparatively simple system. Nevertheless, the accumulation of the nitrogen and the water in the anode channels can lead to a local fuel starvation, which degrades the performance and durability of PEMFCs. In this paper, the behaviors of PEMFCs with a DEA are explored experimentally by detecting the current distribution and the local potentials. The results indicate that the current distribution is uneven during the DEA operation. The local current firstly decreases at the region near the anode outlet, and then extends to the inlet region along the channels with time. The complete fuel starvation near the anode outlet leads to a high local potential and carbon corrosion on the cathode side. The SEM images of the cathode electrode reveal that the significant thickness reduction and the collapse of the electrode's porous structure happen in the cathode catalyst layer, leading to the irreversible decline of the performance. The comparison of the experiments with different oxidants and fuels reveals that the nitrogen crossover from cathode to anode is the dominant factor on the performance decline under the DEA operations.

1. Introduction

Proton exchange membrane fuel cells (PEMFCs) have been considered as a potential alternative to conventional internal combustion engines due to the low operation temperature, fast response, near-zero emissions and favorable power-to-weight ratio. However, several technical issues still prevent the further applications of fuel cells, such as the low hydrogen utilization and the complex system. Under the dead-end anode (DEA) operation, hydrogen is supplied for the reactions on demand by a pressure regulator. Thus, the PEMFCs can obtain high hydrogen utilization. However, when air is employed as the oxidant, the water vapor and inert nitrogen can also crossover the membrane due to the concentration gradients between the anode and cathode [1–4], which results in the local fuel starvation and subsequently carbon corrosion [3,5–7]. Siegel et al. [4] employed neutron images to investigate the liquid water accumulation in a PEMFC with a DEA. Even dry hydrogen was supplied to the anode, the accumulation of liquid water could be still observed in the anode channels in normal operation conditions. The accumulations of water and nitrogen lead to the uneven distribution of the fuel gas. Sasmito et al. [3] and

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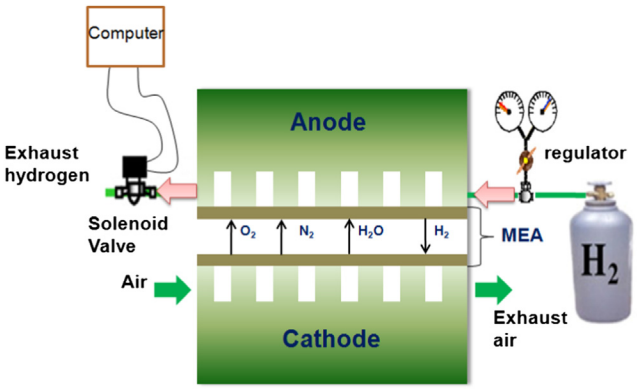

Fig. 1. Schematic of the experimental setup.

Siegel et al. [5] presented models to describe the dynamic evolution of the species in the anode channel and predicted the voltage decline during the purge cycles. Baumgartner et al. [6] adopted a laboratory-scale single cell assembly with four reference electrodes (RHEs) to investigate the electrode potentials under the dead-ended operation. The evolutions of the anode and cathode potentials exhibited that the fuel starvation and carbon corrosion occurred under the DEA operation. The corrosion of carbon support layer was confirmed by the presence of CO_2 in the cathode. Chen et al. [7] developed an along-channel model to predict the carbon corrosion in a PEMFC with a DEA. The model captured the effect of nitrogen accumulation and the current density distribution on the carbon loss, and then predicted the non-recoverable voltage loss. Instructive results were given in the above work, but the irregular distributions of local current density and the corrosion process of PEMFC with a DEA were seldom experimentally investigated.

In this paper, the current distribution, the local potentials, and the catalyst layer micro-structure are measured in order to well understand the PEMFC behaviors under the DEA operation. The degradation mechanism of the PEMFC with a DEA is further analyzed.

2. Experimental

A single cell with the active area of 270 cm^2 was used in the research. Dry hydrogen was fed via a pressure regulator. A solenoid valve was placed at the outlet of the anode for purging control, as shown in Fig. 1. The periodic purging event causes voltage cycling in DEA operation, with a cycle defined as the duration between two consecutive purge events. The tested fuel cell was laid on its side as shown in Fig. 2. A special cathode current collecting end-plate was adopted to measure the current distribution. The end-plate was segmented by embedding fifteen graphite blocks in a plexiglass plate and uniformly arranged in a matrix of 3 × 5. The area of each graphite block was 15.36 cm^2. The segments, which consisted of three parallel graphite blocks perpendicularly to hydrogen flow direction, were also numbered from 5 to 1 along the hydrogen flow direction (see Fig. 2). The flow fields of both cathode and anode were parallel-straight channels. The width and depth of each channel are both 1 mm, with the ratio of channel to rib 1:1 [8].

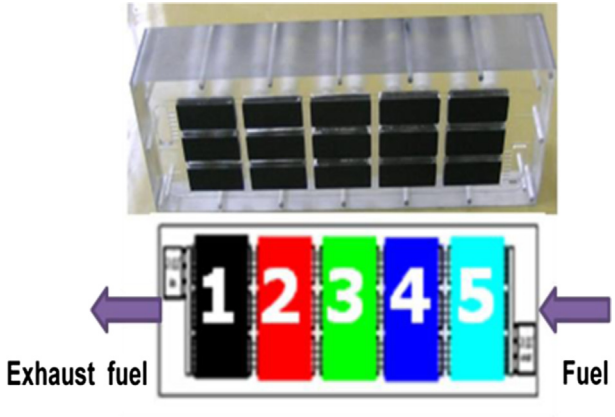

Fig. 2. Schematic of the cathode end-plate.

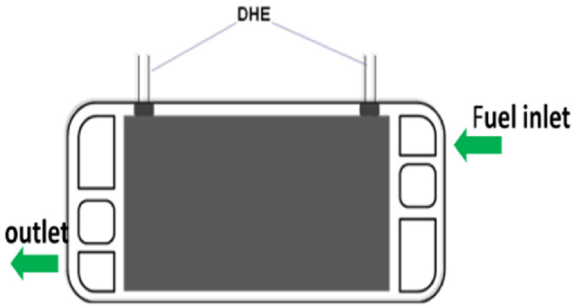

Fig. 3. Schematic of the MEA with DHE.

The dynamic hydrogen reference electrodes (DHEs), which were similar to the design of Siroma et al. [9], were embedded in the membrane electrode assembly (MEA) to measure the local potentials. The DHEs were arranged at the anode inlet and outlet region, as shown in Fig. 3. The local potentials were recorded simultaneously. The applied MEA consisted of five layers: the Nafion® 212 membrane in the middle, and two catalyst layers (CLs) and gas diffusion layers (GDLs) on both sides. The Pt loading of CLs at each side was 0.4 mg cm^{-2}. The GDL included the carbon paper (Toray, TGP-H-060) and a micro pore layer (MPL); and its thickness was about 0.2 mm. In the experiments, the operating conditions of the fuel cells were controlled by the G100 fuel cell test station from Green light Innovation Corp of Canada.

3. Results and discussion

3.1. Local current distribution

The fuel cell interior current distribution was measured by the potentiostatic method. The air flux was fixed at 5.61 L min^{-1}, with

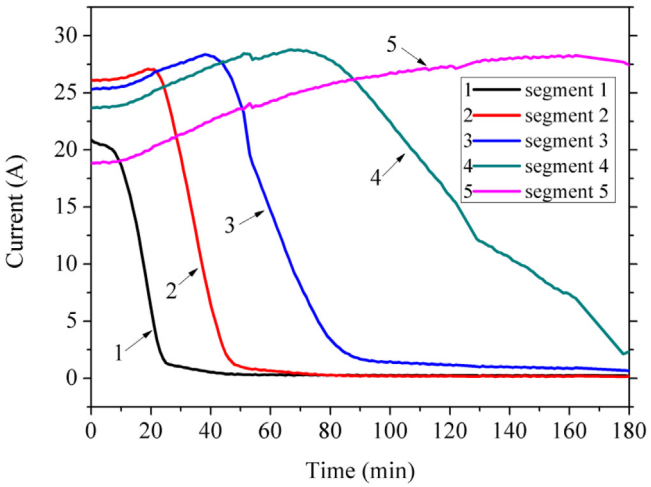

Fig. 4. Evolution of the current distribution.