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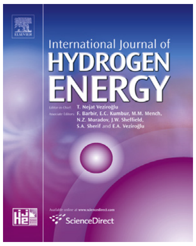

Investigation of self-humidified and dead-ended anode proton exchange membrane fuel cell performance using electrochemical impedance spectroscopy

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ABSTRACT

Self-humidified dead-ended anode proton exchange membrane fuel cell is increasingly being used in some special applications due to its need for simpler and lower cost subsystems. However, the performance of such a fuel cell is more affected by the operational parameters and conditions than the traditional proton exchange membrane fuel cells. Therefore, realizing the most effective parameters and determining their optimum values are essential. In the present study, electrochemical impedance spectroscopy is used to examine the effect of working conditions on the performance of a self-humidified dead-ended anode fuel cell. Working temperature, air stoichiometry, and purge interval are selected to assess their effects on the fuel cell performance. The results show that the performance enhances by increasing the working temperature up to 50 °C, but further increase of the temperature causes an intense reduction in the performance due to a combination of severe membrane drying and build-up of nitrogen in the anode side. The impedance spectra are greatly influenced by the air stoichiometry since increasing the air stoichiometry may lead to severe membrane drying in one hand and increasing mass transport resistance due to accumulation of N_2 in the anode side, on the other hand. While the impedance spectra are less affected by the purge interval at its low values, large values of the purge interval lead to significant mass transport issues. Wasted electrical energy and wasted energy due to hydrogen purging are calculated and compared at different purge intervals.

Introduction

A range of design variants exists within the broad category of proton exchange membrane fuel cells (PEMFCs). Many activities are running in order to lower cost by simplifying PEMFC system and using fewer auxiliary components.

Dead-ended anode (DEA) operation is a common approach as it can simplify the system and requires fewer auxiliary components when compared to the more traditional flow-

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through and circulating anode modes [1–3]. A Dead-ended anode fuel cell is fed by dry hydrogen with regulated pressure at the anode inlet. The configuration usually employs a pressure regulator before the hydrogen inlet to the stack and a purge valve after the anode outlet. Proper water management is crucial in order to keep the membrane sufficiently humidified while ensuring that flooding does not occur in the anode side due to water accumulation [4–7]. In addition, nitrogen can crossover from the cathode side and accumulate at the anode side [8–10]. Therefore, the dead-ended anode operation leads to a gradual voltage loss due to accumulation of water and inert gases in the anode side [3,11]. Hence, the purge valve is periodically opened to partially expel the accumulated liquid water and the inert gases out from the anode side, leading to instantaneous recovery of cell voltage(s) [1].

In another attempt to lower the system cost, necessity of cathode (and/or) anode humidifiers can be eliminated by using a self-humidified membrane in the membrane electrode assembly (MEA). Using of a self-humidified membrane imposes some limitations on the air flow rate and stoichiometry in the cathode side. Since air is not humidified before entering the cathode side of this kind of fuel cell, it has a high tendency to absorb water from the cell and the membrane, especially in the high operating temperatures. Therefore, high air stoichiometry may cause severe membrane drying which leads to performance loss and possibly membrane failure in the extreme conditions. On the other hand, while low air flow rate and stoichiometry may maintain an acceptable level of humidity in the cell, but it leads to lower performance due to lower oxygen concentration at the reaction sites and also may lead to flooding in some cases [12,13].

The combination of a dead-ended anode mode with a self-humidified membrane leads to a very simplified PEM fuel cell system in comparison with traditional systems which require humidifiers, hydrogen circulation pump, and more complicated control system. However, the operation of such a fuel cell is very dependent on the working conditions. It requires careful gas and water management and purge control to achieve optimal operating conditions. For example, finding a suitable amount of air stoichiometry to preserve adequate humidity level in the membrane, appropriate values of purge parameters (close time, open time), and optimum working temperature are the issues that should be addressed in such fuel cells [14].

There are some researches that have developed mathematical and numerical models to investigate the effect of different operating parameters on the performance of the DEA fuel cells. Mokmeli et al. [15] developed a model to simulate the voltage drop of a single cell at the dead-ended anode operation. In their model, voltage loss was attributed to the accumulation of impurity from hydrogen gas. Siegel et al. [16] developed a one-dimensional model to capture the voltage drop in a PEMFC with a dead-ended anode. Sasmito et al. [17] applied the computational fluid dynamic method (CFD) to develop a transient mathematical model to study the effects of operating conditions on the cell performance and nitrogen accumulation in the anode side. Chen et al. developed a model to predict the water and the nitrogen accumulation in the anode channel [18]. Yang et al. [19] developed a model to study the nitrogen accumulation in the anode side of a PEMFC operated in the dead-ended anode mode. Chen et al. [20] employed a dynamic mathematical model to predict the nitrogen accumulation in the anode and its corresponding cell voltage. The model was calibrated and validated using experimental data. A purge strategy based on nitrogen concentration in the anode side was developed by the calibrated model and implemented into controller for anode gas management. Gomez et al. [21] evaluated, experimentally and numerically, the effect of the key operating parameters on the transient performance of a dead-ended anode fuel cell stack. The results suggest that the performance deterioration over time is closely related to the choice of the operating conditions. The study reveals that liquid accumulation at the anode is found to be strongly related to the inlet humidification as well as water transport across the membrane, whereas the cathode stoichiometry affects the nitrogen crossover. Moreover, some studies have been performed to assess the effect of dead-end operation on the degradation processes occurring in the fuel cell [22–24]. However, understanding of what is happening during the dead-end/purge process is a prerequisite for resolving the cause of degradation.

Electrochemical impedance spectroscopy (EIS) is a suitable and powerful diagnostic testing method for fuel cells because it is non-destructive and provides useful information about fuel cell performance and its components without perturbing system from equilibrium. The main advantage of the EIS is its ability to distinguish the individual contributions of the interfacial charge transfer and the mass transport resistances in the catalyst layer and diffusion layer. The disadvantage of this method is that it does not generate local information [25]. EIS provides useful information about fuel cell performance and its components [25–28]. EIS has been used for fault detection and flooding/drying events [29,30], investigating the effect of operating conditions on the fuel cell performance [31].

To our best knowledge, there are a few numbers of researches which used EIS for dead-end/purge analysis due to the challenge of capturing the process on the relatively short duration of the purge event. Meyer et al. [1], in an attempt to resolve this issue, used single-frequency/high-frequency impedance measurements, along with a ‘reconstructed impedance’ method that combines the results of consecutive repeatable cycles to build up full EIS during through flow/dead-ended operation. They used EIS to examine the source of performance decay during dead-ended operation. Their results showed that there is an increase in performance at initial stages of dead-ended operation caused by improved hydration of the membrane electrolyte and increased anode compartment pressure. An increase in mass transport losses due to a combination of accumulation of N_2 in the anode and water management leads to subsequent reduction in performance.

Strahl et al. [3] used fast EIS to study the hydrogen purge effects on performance and efficiency of an open-cathode DEA PEMFC system. Their results showed that the major DEA operation performance limitation is related to water instead of N_2. Lee et al. investigated the dynamic behavior of a self-humidified PEMFC single cell by the EIS method, although, their single cell was not operated under DEA conditions [32].

In the present study, we use the EIS test method to investigate the effect of different operating parameters including

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